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ABSTRACT

Pollution of the general environment, which exposes an entire population group for an indeterminate period of time, certainly constitutes a problem in public health. Serious aid pollution episodes have resulted in increased mortality and a possible relationship between chronic exposure to a polluted atmosphere and certain diseases has been suggested. Thus, this monograph brings together a body of recent information on a number of aspects of air pollution for the benefit of public officials, and in particular public health officials, who are faced with the necessity of taking appropriate and effective action. Essays are titled: A Historical Review of Atmospheric Pollution; The Identification of the Air Pollution Problem; The Role of Meteorology in Air Pollution; Sampling, Analysis and Instrumentation in the Field of Air Pollution; Some Aspects of the Physical and Chemical Nature of Air Pollution; Effects of Air Pollution on Human Health; Effects of Air Pollution on Animals; Effects of Air Pollution on Plants; Economic and Social Aspects of Air Pollution; Control of Air Pollution by Site Selection and Zoning; Prevention and Control of Air Pollution by Process Changes or Equipment; Fuel Selection and Utilization; Air Pollution Legislation: Standards and Enforcement; and Radioactive Pollution of the Atmosphere. (BL)

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Introduction

The problem of air pollution has existed for centuries. Smoke, ash, sulfur dioxide and other products of simple combustion have long been recognized as a nuisance, and a costly one. At the present time, the belief that air pollution not only is a source of discomfort but also may constitute a menace to human health has grown to a conviction. Serious air pollution episodes have certainly resulted in increased mortality, and independent investigations have suggested a possible relationship between chronic exposure to a polluted atmosphere and such diseases as acute bronchitis and primary lung cancer.

There is a distinction between the pollution of air in a factory or other occupational environment, where the workers—usually healthy adult males—are exposed only during their working hours, and the pollution of the general environment, which exposes an entire population group for an indeterminate period of time. Such a general exposure constitutes a problem in public health, and is properly a matter of concern to public health authorities.

The control of air pollution is no longer concerned merely with the products of simple combustion of conventional fuels. Industrial effluents discharged into the atmosphere are increasing in amount, in diversity and in complexity. As a consequence of developing industrialization, of the expanding use of internal combustion engines, and of the greater densities of population exposed to air pollutants, the seriousness of the situation is rapidly increasing, and the difficulties of applying effective controls are becoming progressively greater.

This monograph brings together a body of recent information on a number of aspects of air pollution for the benefit of public officials, and in particular public health officials, who are faced with the necessity of taking appropriate and effective action. In order to ensure that the material included herein faithfully reflected world trends in air pollution research, the individual chapters were submitted to a panel of experts in the field, five of whom contributed to the monograph. These authorities, who attended a meeting of the WHO Expert Committee on Environmental Sanitation in 1957, are as follows: Dr E. C. Halliday, Head, General Physics Division, National Physical Research Laboratory, Council of Scientific and Industrial Research, Pretoria, Union of South Africa; Dr Harry Heimann, formerly Chief, Operational Research Section, Air Pollution Medical Program, US Public Health Service, Washington, D.C., USA; Dr E. Leclerc, Professor of Industrial Chemistry and Sanitary Techniques, University of Liège, Belgium; Dr Louis C. McCabe, President, Resources Research Inc., Washington, D.C., USA; Dr Albert

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Thanks are due to Professor L. Silverman of Harvard University, who re-examined the material and gave valuable guidance on technical aspects.

E. C. HALLIDAY, M.Sc., Ph.D. *

A HISTORICAL REVIEW OF ATMOSPHERIC POLLUTION

Introduction

Man cannot live without breathing oxygen diluted with some inert gas, and in the atmosphere there is a suitable mixture of oxygen and nitrogen. But in addition there is a variety of gases, vapours and aerosols, varying in content and concentration at different places over the surface of the earth, and these he needs must breathe in too. Some of the materials in question are physiologically inert, but others produce reactions which range from mild inconvenience to severe toxicity.

A simple consideration indicates that there has never been a truly unpolluted atmosphere, for decaying vegetable matter, decaying dead animals and the products of forest fires have emitted gaseous and particulate matter since the world began. However, atmospheric pollution, as a subject for concern on the part of the general public, has come into existence only as the result of technology. The first technical development of man was the use of fire, which produced carbon dioxide, smoke and ash; and the atmosphere of all towns, even in early civilizations, must have contained a considerable amount of pollution. Accounts of mediaeval towns in Europe indicate that the air was fouled not only by wood smoke but also by the odours of many domestic activities, of noxious trades such as tanning, and of the decaying rubbish in the streets caused by a most primitive sanitation system. Nevertheless, the citizens of the towns do not seem to have been unduly concerned by the quality of the atmosphere which they breathed.

With the introduction of coal as a source of heat, signs of the rise of public dissatisfaction are noted by historians, and one might say that atmospheric pollution as a social problem dates from the beginning of the 14th century. But a scientific and technical review of the history of air pollution cannot commence much before the year 1850, for before that time, though there was much talk about the obnoxiousness of smoke, little knowledge was available and so no progress was made in the control of pollutants.

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An over-all view of the hundred years since the first steps were taken in understanding the nature of air pollution indicates that two processes have been in operation. On the one hand, developing techniques have introduced new forms of pollution into the atmosphere and, on the other, town populations have become more intolerant of the types of pollution which they were having to breathe. With this growing dislike of pollutants, interest has spread to the effects of these materials not only upon man himself but also on vegetation and animals, upon buildings, clothing, works of art and other articles of property. This latter development is hardly within the immediate scope of public health, and yet the study of atmospheric pollution can scarcely fail to take notice of it, since it indirectly affects man's mental, if not physiological, well-being.

It therefore appears that atmospheric pollution consists of that material, gaseous or particulate, which is commonly found associated with the oxygen and nitrogen of the atmosphere and which is (*a*) toxic, (*b*) irritant, and (*c*) in the nature of a hardship to man, either directly or because of its toxic or harmful effects upon animals, vegetation or human property.

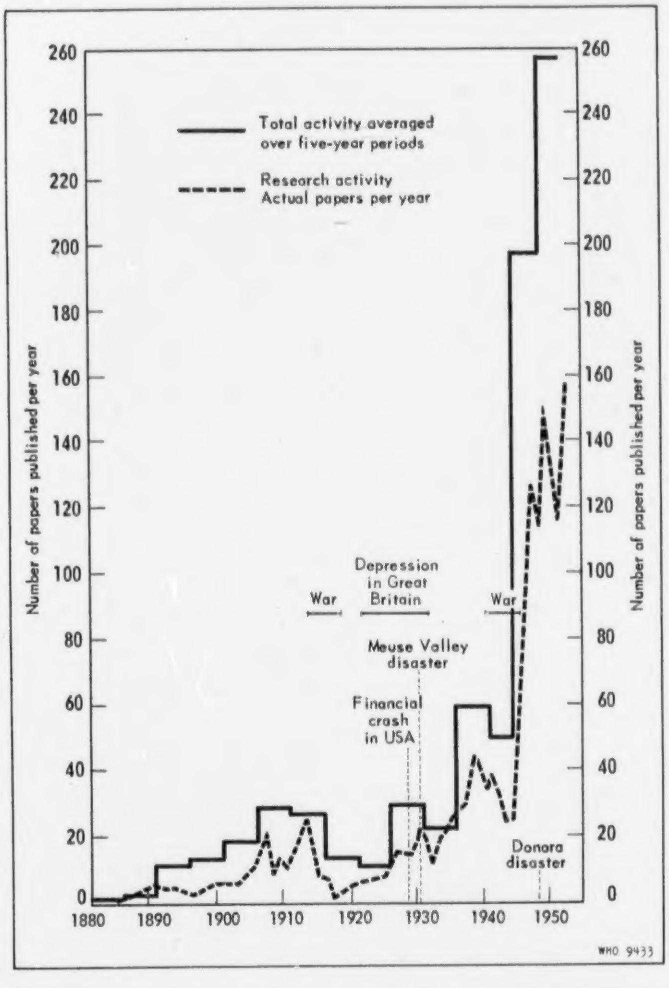
In the time available for the preparation of this historical survey it has not been possible to make a very full study of the many scientific and technical publications which have marked the progress in the study and control of pollutants in the air. Considerable use has been made of (1) a bibliography prepared by the US Bureau of Mines (Davenport & Morgis, 1954); (2) a bibliography attached to an article by Schwartz et al. (1955); (3) a bibliography drawn up by the Fuel Research Station of the Department of Scientific and Industrial Research (Great Britain, Department of Scientific and Industrial Research, 1956); (4) a monograph on air pollution (Meetham, 1952) and a book on smoke (Marsh, 1947), a compilation of papers on air pollution (Mallette, 1955) and a number of reports which have been sent by public health departments or prominent research workers in France, the Netherlands, Austria, Italy and Australia.

It was thought that graphic demonstrations of the growth of activity in the field of air pollution might help in an appreciation of the subject, and for this purpose the US Bureau of Mines bibliography was made use of, although it was clear that in this volume the information about work in Germany and France was very inadequate, and that quite a number of articles published in England had escaped attention. In addition, the bibliography lists some papers more than once if they happen to deal with several of the subjects in the classification adopted by the compilers. Nevertheless, it was thought that in general the bibliography was a complete enough collection of data on which to construct graphs that would give a revealing picture of historical development.

The first graph (Fig. 1) shows the development of total world activity in the form of a histogram based on five-year averages. Superimposed on the histogram is a graph of the actual number of research papers published every year for the same period. The second graph (Fig. 2) shows the development of world activity under three headings—the nature and origin of

FIG. 1

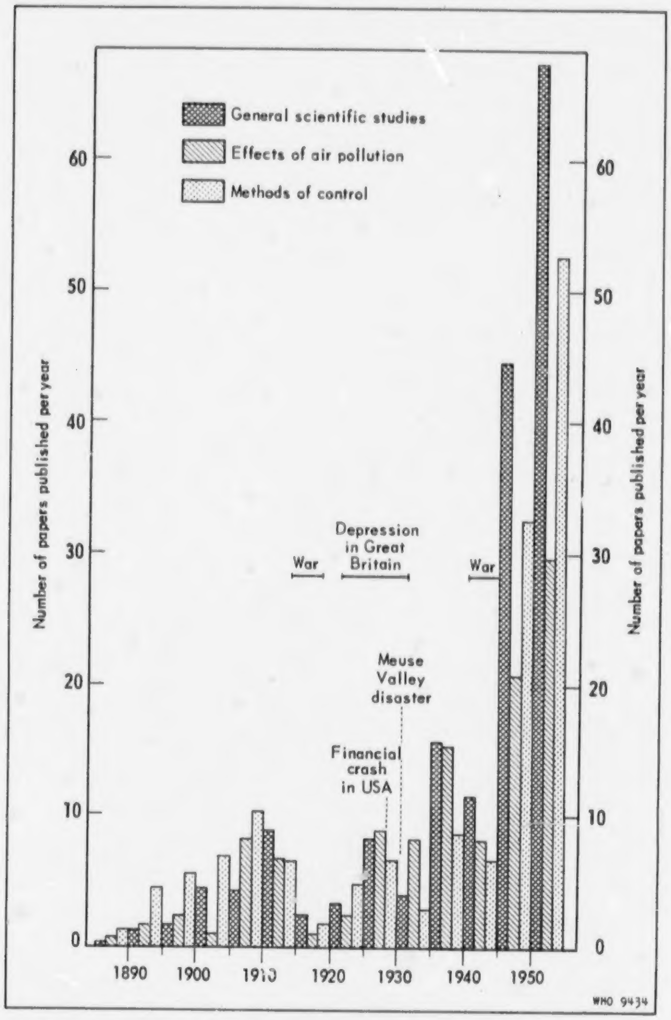
PROGRESS OF TOTAL TECHNICAL ACTIVITY AND OF RESEARCH ACTIVITY IN THE FIELD OF ATMOSPHERIC POLLUTION



pollution, effects of air pollution (medical and other), and the methods of control. These graphs show that (*a*) there has been a steady development in

FIG. 2

DEVELOPMENT OF THREE DIFFERENT LINES OF INVESTIGATION IN THE FIELD OF ATMOSPHERIC POLLUTION



The three histograms give average figures for five-year periods.

Though each column covers a space of about one year on the time axis, it actually applies to the whole of the five-year period in which it stands.

scientific and technical activity in the field of air pollution for over sixty years; (*b*) though medical and technical activity led the way prior to 1900, physical and chemical research is now playing a leading part in the studies; (*c*) activity in the study of air pollution has been adversely affected both by wars and by economic depressions, probably more than other forms of scientific investigation. This indicates that air pollution research has been a marginal activity, a sort of " extra " to the normal community interests, and has not achieved the status attained by other branches of public hygiene (such as food pollution and epidemic control); and (*d*) activity in air pollution studies has increased during the last ten years at a very much faster rate than during the previous fifty years, the slope of the graphs indicating that at least one strong new influence must have made itself felt about 1945. It is not possible to continue the graphs beyond 1952 because knowledge of the papers that have been published during the years 1953-55 is incomplete. The card index which the US Library of Congress is compiling is by no means complete, so that it would not be a true indication of the present state of affairs to make use of the information at present available, but the general impression gained from a study of the various journals is that the rate of publication and the amount of study being carried out are still increasing.

The Different Pollutants and their Place in History

Smoke is the first pollutant to have attracted community attention, and its history goes back to the 13th century. The immediate cause of the production of smoke in such quantities as to constitute a nuisance was the exhaustion of the supplies of wood fuel in Europe and the introduction of coal as a substitute. The growth of populations and the development of manufacturing methods produced an increased demand for fuel, rapidly depleting the reserves of wood in the afforested areas. In very many articles which have been written on the subject (Davenport & Morgis, 1954) reference is made to the conditions in London as early as 1300, when a royal proclamation prohibited the use of coal in London, and from that time onwards communities have suffered from smoke and soot, until comparatively recently without relief.

Sulfur dioxide is the second pollutant to cause community discomfort, for it is produced at the same time as smoke, from the burning of coal; but for quite three hundred years the sulfur dioxide was not recognized as a separate pollutant because of the inadequate chemical knowledge available. All that was known was that smoke was accompanied by an unpleasant smell and an irritation to the throat and nose. By 1600, however, it was well known that the sulfur in the coal was responsible for the annoyance, and methods of coking coal to remove some of the sulfur and volatile components were already being developed.

Sulfur dioxide in much higher concentrations than are met with as a result of the combustion of coal has become a pollutant wherever the metallurgical industry has developed, for so many metallic ores are sulfides that a very high percentage of processes for producing pure metals also produce tremendous quantities of sulfur dioxide.

Hydrochloric acid first became a recognized atmospheric pollutant some time after 1800, with the development of the chemical industry. In the production of sodium carbonate from common salt a strong emission of hydrochloric acid was produced which did great damage to property and vegetation.

The chemical industry also introduced such pollutants as hydrogen sulfide from tar distillation, nitrogen dioxide from the chamber process for sulfuric acid, hydrogen fluoride from the production of super phosphate fertilizer (and later from the manufacture of aluminium), while the metallurgical industry introduced the community to a variety of poisonous or noxious fumes from such metals as lead, arsenic, zinc and copper. In very recent times the use of beryllium for a number of industrial purposes has created a fresh hazard in the form of finely divided beryllium, and there is a likelihood that in the future new industrial techniques will bring with them their own types of pollutant, which will first be experienced by the industrial worker and may later become part of the pollution of city air unless steps are taken to control the processes.

From what has been said above one fact emerges which must always be remembered—that pollution of the air of the city is an extension of the pollution of the air of the factory, so that the science of air pollution control is an extension of the science of industrial hygiene. The methods of measurement are similar in principle but not in practice, for the industrial hygienist is working in a semi-closed atmosphere where concentrations are higher than they are in the street, and his conditions are slightly simpler, so that sampling processes which last for only a few minutes are usually sufficient to give him a picture of what is taking place in the factory, while sampling procedures in the street must usually continue for months and often for years before an understanding is obtained of the processes concerned in the production of the pollution. Thus the new forms of pollution which have been encountered in Los Angeles since 1945 are still not well understood, although measurements have been proceeding for nearly ten years, virtually without interruption. These pollutants cannot be named in this outline because their nature is not yet properly understood, but it is quite clear that they are a type of pollutant which can be expected to play an increasingly important part in the atmospheres of cities in the near future.

The History of Smoke and Sulfur Dioxide

Until perhaps 1940 or thereabouts the words " air pollution " meant for most people smoke and sulfur dioxide. A study of the bibliographies which have been prepared on the general subject of atmospheric pollution (Davenport & Morgis, 1954; Great Britain, Department of Scientific and Industrial Research, 1956; Schwartz et al., 1955) indicates that almost all the scientific and technical papers which were written before that date deal with smoke, its manner of production, its effect, methods of determining the emission rate from stacks, and technical methods of reducing the emission of smoke from industrial furnaces and of lowering the concentration of sulfur dioxide in the neighbourhood of refineries and smelters. So the problem of smoke and sulfur dioxide far outweighed in interest the problems produced by any other form of pollution.

The Bureau of Mines bibliography lists only thirty articles, on the nature, composition and effects of air pollution, published in the thirty years before 1890, and all of them are concerned with the effects of combustion. Not a single paper was written upon the technique of determining the nature and concentration of this pollution. On the other hand, nearly eighty articles are listed on the subject of the control of air pollution by means of stoking techniques and the use of smokeless fuels. This shows the small part that scientific methods and the scientific mind were taking in the handling of this problem.

For the following ten years the bibliography lists thirty-three articles on the composition and effects of pollution and eighty-eight on methods of control, but still nothing had been written on the technique of the determination of concentrations of pollutants. Practically all of this literature was concerned with smoke and sulfur dioxide, although, as has been noted earlier, the production of hydrochloric acid and hydrogen sulfide pollutants had already developed considerably as a result of the growth of the chemical industry. However, the fact that smoke was an almost universal blight upon the city populations of the United States of America, Great Britain and other countries of Europe caused it to claim almost all technical interest. As early as 1880 an article in the *Journal of the Royal Society of Arts* (Carpenter, 1880) stressed that coal combustion in low-temperature domestic and semi-domestic furnaces was a major cause of smoke, while in 1897 the *Journal of the Franklin Institute* analysed very concisely the mechanism of coal combustion and indicated that low-temperature combustion is a prolific source of smoke; and in 1899 a lecturer in London (Beilby, 1899) commented on the same factors, condemned over-stoking, inadequate air supply and low-temperature combustion, discussed the need for mechanical stokers and smokeless fuels, and showed that smoke is the result of abuse of combustion equipment. Thus by the turn of the century almost everything that is known today about the causes of smoke and their elimination had already been said, but hardly anything had been done to reduce the smokiness of cities.

The reason for this inability to take advantage of knowledge had been stated very clearly by a writer in a British building journal (*Builder (Lond.)*, 1899). He had pointed out that (*a*) smoke was a by-product of an activity which commanded the attention and the support of all financial interests in the country because it produced goods and profits, and therefore the willingness to give thought to the smoke aspect of the activity was very limited; (*b*) the damage done by smoke, though very considerable throughout the country, was not very clearly visible to the individual smoke producer, because it was very widely spread, so that interest in the reduction of smoke was lacking; and (*c*) since the damage done by smoke was due to a very large number of small producers of smoke, a clear relationship between cause and effect was difficult to establish.

This analysis of the situation remains true to this day. The control of smokiness depends upon the control of the production of smoke, and the control of a very large number of sources is very inadequate when the owners of the sources do not see any clear need for such control in their particular case, even though they may be quite convinced of the undesirability of the smokiness as a whole.

The history of smoke control in American cities, such as St Louis, Pittsburgh, and Cincinnati, shows that even when a very large volume of public opinion was convinced of the need for smoke reduction, that same public was militantly unwilling to take the technical steps necessary for the reduction, the mental attitude always being that the responsibility for cure must surely lie at someone else's door. The following quotation comes from a historical article by R. R. Tucker (1945), one-time Smoke Commissioner to the City of St Louis:

> In 1925 a fresh start was initiated . . . the attempt was to be one wherein the general public would be educated to the advantages of the elimination of smoke. A competent staff was employed . . . A firing school was started in the center of the city and instructions were given to individuals in the proper method of firing typical equipment used in St Louis. While in progress a survey showed that there was a material reduction in the quantity of smoke discharged into the atmosphere. *This was not however readily apparent to the average citizen.* As the campaign tapered off interest waned and St Louisians returned to their slovenly habits of firing.

With reference to the 1939 smoke regulations, he writes, " Enforcement of the ordinance was resisted by some, and brochures were even distributed within a radius of two miles of the Commissioner's house insinuating that the Commissioner was dishonest and was showing favouritism for monetary considerations. "

During the period 1890-1914 quite a number of studies were made of the smoke situation in various cities. A very intense investigation was conducted in Chicago in about 1910, resulting in the publication of a printed report of some five hundred pages in which almost everything that is studied nowadays regarding smoke, soot and ash was examined and reported upon. Other cities investigated were St Louis, in 1907 (*League Amer. Munic. Bull.*, 1907), and Pittsburgh, in 1912 (Pittsburgh University, 1912). A large scale measurement of smoke, sulfur dioxide and soot-and-ash fall was started in Great Britain in 1912 (*Sci. Amer.* 1914a), and has continued for over forty years, with a steady increase in the number of measuring stations. (In passing it is interesting to note that the possible origin of the British " lead peroxide method " of estimating sulfur dioxide lies in a paper by Witz in 1885 on the smoke of Paris, in which it is suggested that the attack of sulfur dioxide on the lead peroxide in paint might be made a method of studying the pollution of the atmosphere.)

In spite of these studies and even in spite of the passage of a number of ordinances in Cincinnati, Montreal, Chicago, St Louis, Pittsburgh and other cities, and of laws in Germany (Schwartz et al., 1955, p. 542) and Italy (Mallette, 1955, p. 253), the smoke position apparently did not improve, for in 1930 city agitation on the matter of smoke was still an issue. By this time it was becoming apparent not only that the remarks made in the *Builder* (1899) and on various subsequent occasions were true, but that as the small consumers of coal had no really effective method of preventing the production of smoke from their small and inefficient appliances, legislation and penalties were of almost no effect, so that the only way of stopping smoke at the source was to supply a fuel which was very nearly incapable of producing smoke. As a result cities such as St Louis and Pittsburgh prohibited the sale of bituminous coal with a volatile content of more than 20% and provided alternative smokeless fuels such as anthracite, or processed smokeless fuels such as Pittsburgh's " Disco ". At the same time the increasing supply of oil or of natural gas as a heating fuel in many American cities, at a price comparable with that of coal, caused a widespread switch to these fuels and in this way dramatically changed the pollution situation in many of the great cities of the USA.

Great Britain was in a vastly different situation, for natural gas was unobtainable, and oil had to be imported and paid for with foreign exchange, so that coal gas, coke and smokeless solid fuel were the only substitutes available. At this point, economic depression and then war intervened to prevent action in Great Britain. It is fairly certain that the programme of pollution measurement which was started in 1912 was intended to be a preliminary to a programme of smoke abatement. However, the 1914-18 war held up the measurement programme, and by 1925 the economic situation was such that no one had the heart to urge any action that did not have the promise of increased earnings as its immediate goal. Then came rumours of war and finally war itself, and it was not until 1946 that the country was once again in a position to consider the situation. By that time it had become clear that with the high price of coal and the reduced coal reserves of Great Britain, even that saving in coal which would be obtained by a smoke elimination campaign would be extremely valuable, apart from the other advantages to be expected from smoke reduction. With the country in this frame of mind it was possible for national action to be considered, and this led to the Clean Air Act, which is now in its early stages of application. The Act is based upon the same realization that motivated the anti-smoke administrators in the American cities—that only a rationalized use of fuel would lead to the elimination of smoke. The Act therefore involves considerable national planning (*a*) to equip all new houses, and by degrees all old houses, with equipment which will burn smokeless fuel—either gas, coke or low volatile carbonized coal; and (*b*) to develop throughout the country an efficient use of the country's coal for the purpose of supplying gas, coke, smokeless solid fuel (by several different methods suitable for different raw supplies) and such by-products as benzol, pharmaceutical and other chemical raw materials, road tar and fuel tar for large boilers. This process has only recently been started and it will be intensely interesting to watch its development.

It is probably worth while to consider briefly the factors which have caused the Americans to apply city control and the British to apply national control. In both cases the clue to the solution is rationalized fuel supply. In the USA unorganized processes, such as the development of waste natural gas supplies and of oil burners for house heating, at a price competitive with coal, caused householders and others to turn to the right sort of fuel for the purpose of reducing smoke emission. In addition, the USA being a federation and extremely large in area, with an expanding economy and fiscal funds in plenty, the conditions were right for city control of smoke emission, aided by state legislation to give the cities the necessary powers. In Great Britain the question of fuel supply was difficult to solve and, in addition, by the time that public opposition to air pollution had grown strong, the economic situation had become stationary, leaving very few surplus funds to allow for independent ventures by cities. Thus, although under the terms of the Clean Air Act the cities and local authorities are entrusted with the carrying out of many provisions of the Act, the drive for the elimination of smoke has come from the central government, which alone can sponsor a study of the fuel situation and its rational development in the direction of the most economic supply of smokeless fuel.

In all this one must not lose sight of the part played by the smoke abatement societies in both countries, for the problem being faced was that of changing the habits of a nation, and only considerable and capable propaganda activity could develop the thinking of citizens sufficiently to make it possible for these administrative programmes to receive satisfactory support. There is no great value in tracing the rise and fall of various smoke abatement associations in the USA and Great Britain, but reference should be made to the two main associations, the Air Pollution Control Association in the USA, which not long ago celebrated its fiftieth anniversary, and the National Smoke Abatement Association in Great Britain, whose journal *Smokeless Air* has recently published its hundredth issue. These associations, by public meetings, propaganda literature, appeals to the public, the industrialists and the government, and by other activities, have continually stressed the need to take atmospheric pollution seriously and not to allow it to become a chronic condition of city life.

Up to this point nothing has been said about sulfur dioxide in association with smoke because, while the emission of smoke is difficult to prevent, the emission of sulfur dioxide when coal is burnt is almost impossible to prevent. Some of the 1%-4% of sulfur which is present in coal can be removed by washing the heavy pyrites from the lighter coal, but some sulfur is always left and the result is the production of sulfur dioxide when the coal is burned. If the concentration of sulfur dioxide in the flue gases were high the chemical extraction of the sulfur might be possible, but the very large volumes of gas which have to be treated make the processes very intractable. Two large electricity plants in London have installed flue-gas scrubbing equipment in spite of the very high costs involved (Meetham, 1952, p. 239; Pearson, Nonhebel & Ulander, 1936), but nowhere else in the world have industries using large quantities of coal been able to face the problem of sulfur recovery. Large industries produce little smoke but large quantities of sulfur dioxide, so this is their main problem and it is far from easy to solve. Small consumers of coal make much more smoke and much less sulfur dioxide, so that their problem is efficient combustion, which is capable of solution.

Smoke and Sulfur Dioxide in Europe

Pollution by smoke and soot in the continent of Europe has had a history somewhat similar to that in Great Britain and the USA, but with variations due to local conditions. In Germany, the Netherlands and Austria the domestic pattern of fuel usage in many cities is the use of gas for cooking and for water heating and the use of coke for space heating, in box-type stoves. This domestic pattern produces hardly any pollution, so that industrial sources begin to be of predominant interest. Nevertheless, measurement in Berlin in 1954 (Heller, 1954) showed quite a high proportion of smoke of domestic origin. Towards the end of the 19th century there is evidence of extensive damage being done to forests in Germany by the sulfur dioxide from smelters, and even to this day the heavily industrialized Ruhr Valley is an area of intense smoke pollution, from both industrial and domestic sources. But in general it may be said that the conditions in Germany, the Netherlands and Austria were better than they were in Great Britain and the USA. One writer has said that this is not so much due to a consideration of the need for the reduction of air pollution as to a need to conserve every ton of fuel available. Nevertheless, in 1928 a writer claims that the public health authorities should give more consideration to the control of smoke and fumes (Hahr, 1928), and in 1943 another writer speaks of the necessity for investigations on the nature of damage to plant life due to waste gases (Koritnig, 1943).

The legal situation is that in Germany and Austria laws were passed, in 1820 and 1909 in the former country and in 1811 in the latter, with the object of enabling any person who suffered damage through the action of waste gases upon himself, his livestock or his property to claim compensation. The result of this has been that industries have had to watch their effluents because of the possibility of large claims for damages, and as a result associations of industrialists, such as the Vereinigung der Grosskesselbesitzer, discuss at their conferences technical papers on the prevention of smoke emission; in addition the literature includes a large number of articles on methods of assessing damage to plants and property. The laws, however, make no provision for damage caused by the effluents from domestic chimneys, and there is at present a move in Germany to bring about the adoption of legislation which will control the emission of smoke and fumes from domestic buildings and from vehicles. The situation in France seems to be somewhat similar. Legislation was passed in 1932 (Schwartz et al., 1955, p. 540) defining what shall be tolerated in the way of smoke and fume production, and the implementation of this legislation has been left in the hands of the local authorities. Such pollution as is caused by domestic combustion is uncontrolled, but the use of coal in France is not as extensive as it is in Great Britain, so that conditions on the whole are not severe. However, measurements of smokiness and of sulfur dioxide concentration in Paris during 1950-56 show that the smokiness of Paris is comparable with that of London on certain occasions, and a government commission was appointed in 1954 to consider the dangers which are caused in the country by the emission of pollutants to the air (Raymond, 1956).

Italy has become a city-dwelling industrial country only comparatively recently, and even now almost all its southern portions are largely farming lands. A technical article (Mallette, 1955, p. 252) states that, owing to the meteorological conditions of Italy, the accumulation of pollutants is less than it is in other parts of the world, but gives quite a number of cases of physiological upset due to pollutants, indicating that there must often be conditions in which dangerous concentrations of pollutants are present.

Italian law on air pollution, dating from 1912, seems to be concerned only with the control of effluents from industries, so that one is inclined to say that public opinion in Italy, of a nature unfavourable to pollutants in the air, has grown rather slowly.

General Review of the More Specific Pollutants

The sources of air pollution from the products of combustion are almost always very numerous and widespread. This type of pollution is therefore diffuse, the concentration levels usually varying only slightly from place to place in a city. The air pollution caused by the emission of specific pollutants, such as SO_2 from smelters, HCl and NO_2 from chemical industry, HF from aluminium plants and fertilizer factories and some foundries, H_2S from gas retorts, and arsenic, lead and zinc fumes from some metallurgical processes, has this characteristic that the number of such sources in or near a city will usually be limited and the affected area around each source is usually small. Thus the offensiveness or the toxicity of the effluent is found in a confined area, and the complaint rate is consequently lower, while the source of the effluent, once it has been identified by techniques (which have been developed in the practice of industrial hygiene and are now used in the control of city air pollution) is fairly easily discovered. The result is that the problem of these pollutants is actually not as difficult as that of the products of combustion, and less has been written on the subject.

In the Bureau of Mines bibliography up to the year 1930 nearly 90 per cent of the articles listed were concerned with the products of combustion and only 10 per cent dealt with specific pollutants. After 1945 only 45 per cent of articles were concerned with products of combustion, 20 per cent discussed aerosols, and 22 per cent dealt with the toxic and noxious chemical gases. The cause of the heightened scientific and technical interest must to some degree be due to the increasing activities of city pollution-control organizations, which had been covering the chemical industries as part and parcel of their normal activities throughout the USA. These control organizations were in all cases originally established to handle the products of combustion, but as soon as their activities had become streamlined they became conscious of the localized pollution situations caused by the chemical and metallurgical industry and included them in the control scheme.

The administrative pattern for the control of such pollutants in the USA has been for each city to draw up its own regulations and specifications for the concentrations of pollutant (varying from type to type) which might be allowed to escape from a stack. Some of the states have passed basic legislation giving the cities a set of principles on which to work, while other states have taken each application made by each city for powers to control pollution on its own merits and approved or modified the terms of the application as the situation seemed to require (Smoke Prevention Association of America, 1922).

In the continent of Europe the pattern of action has on the whole been to pass national legislation specifying what concentrations of different pollutants would be tolerated, and to leave the application of the law to local authorities. One interesting development was the early action of Italy—in 1912—in classifying a group of industries as dangerous and prohibiting their operation at less than a stated distance from a town or city. This of course left the country folk to bear the uncontrolled effluents; and, moreover, as time has passed, some of these industries have been enveloped by the towns, so that the legal provisions have become inoperative.

In Great Britain a rather different approach was adopted when the chemical industry began to develop in the second half of the 19th century. The emissions of strong acid fumes from the new factories were very offensive and dangerous for quite a distance round each factory, and the first reaction of government authorities was to forbid the operation of the factories altogether. However, the commercial importance of the activities concerned to the country's economy was realized, and so under the terms of the " Alkali, etc. Works Regulation Act ", first passed in 1863 and amended in 1878, in 1881 and in subsequent years, all these factories were placed under the care of a specially created government-controlled inspectorate.

The object of the inspectorate was to ensure that all these factory processes were operated at the highest level of efficiency known to the chemical industry. With two exceptions, no statutory specifications were written for the maximum amount of effluent which would be allowed from any given stack. Instead, the inspectors acted as advisers and consultants to the industries under their care, and saw to it that any discovery of a method of reducing effluent in one factory was immediately communicated to all similar factories and put into operation there. This method of control is not directly based upon a health criterion, but upon a " chemical efficiency " conception, so that the " Alkali Inspectors " as they are called are all men of considerable experience in chemical industry rather than in industrial hygiene or sanitary engineering, but the effect of the control method has been to produce steady reductions in effluent levels in chemical factory stacks. In addition, this line of action has fostered the development of quite a number of new uses for pollutants scrubbed from industrial effluents, so that waste has been turned into a marketable by-product.

It should be noted that although the organization of the prevention of chemical effluent emission in the USA had its origins in public hygiene, the actual basis of operation of the control authorities in many American towns is almost identical with that of the British Alkali Inspectorate. It has been found that the inspector who considers a factory merely from the hygiene aspect can only attempt to control the effluent escaping from the factory; but since he is not an industrial chemist he cannot enter into a consideration of the processes in the factory which produce the effluent, and so cannot be of any assistance to the industrialist in advising him as to the technique to be employed in reducing the effluent, or, on occasions, the method to be used for turning mere waste into an income-producing by-product.

" Los Angeles " Pollution : a New Type

In 1943 Los Angeles began to experience a new type of pollution which has caused the city to become known throughout the world for its smog. It is now thought that similar pollutants are to be found in almost all large cities, but Los Angeles was the first area to experience high concentrations of this new pollutant, because of two circumstances more marked there than in most other cities of the world. Los Angeles has a poor natural ventilation (on a meteorological scale) and it also uses no coal but has a very high consumption of petrol. The result is that the atmospheric concentration of sulfur dioxide is very low, but because of the stagnation of the air over the city, other pollutants, which are now thought to be of a petroleum origin, are held in the air-volume above the city until quite high concentrations result.

Los Angeles became a very highly industrialized area during the recent world war, and its population increased very rapidly. By 1944 the inhabitants became aware of an atmospheric condition which appeared and disappeared, but sometimes remained for days at a time, in which visibility was reduced markedly by a light blue haze, and many persons suffered from sore throat, running nose and eyes, and varying degrees of headache. Air pollution officials from the eastern towns of the USA came to study the situation and decided that the cause was sulfur dioxide, which can produce some of the above symptoms, though usually not all of them. A programme for the reduction of sulfur dioxide emission by various industries, including the oil refineries, did not produce the expected reduction in the smog, as it was called, so further investigation was made and it was discovered that petrol vapour (hydrocarbons) in combination with perhaps nitrogen dioxide or some other reagent present in the air, and under the influence of the energy of sunlight, produced some organic compound which has still not been defined with certainty but which is thought to be a strong oxidizer. A substance which was made from hydrocarbons and nitrogen dioxide, irradiated by ultraviolet light under experimental conditions, was shown to possess the lachrymatory properties of Los Angeles smog, and so the smog control agency set out to prevent the escape of petrol vapours from the storage tanks of the oil refineries. This activity still did not have the required effect, so further thought was given to the subject and it was realized that the very high number of two and a half million motor cars in Los Angeles used five million gallons of petrol a day and, because of the inefficiency of the automobile carburetter, emitted over a thousand tons of hydrocarbons to the atmosphere daily. This means that in a new situation the old problem has again arisen—namely, that the major source of the pollution comes from a very large number of domestic consumers of fuel, this time petrol. As in the case of domestic coal combustion in St Louis and Pittsburgh, the cure cannot be found in terms of legislation forbidding motorists to produce hydrocarbon effluent. It must consist in technical advances which will enable the automobile engine to burn its fuel with an efficiency approaching 100 per cent. This is the present situation in Los Angeles, whose short history of ten years of air pollution research has made air pollution officials in all the large cities of the world reconsider their position for the future. If hydrocarbons from automobile exhausts are the cause of a major problem in Los Angeles, then the problem can appear in other cities as soon as the number of automobiles becomes large enough, and in all cities the number is steadily rising. Scientific articles on Los Angeles smog are very numerous, but luckily a good many of them are gathered together in the proceedings of conferences which have been held on the Los Angeles situation (California State Department of Public Health, 1955; *National air pollution symposia*, 1949, 1952, 1955; Stanford Research Institute, 1954).

History of the Scientific Analysis of Air Pollution

The following is a short review of the scientific aspect of the study of air pollution, leaving out almost entirely the technical and engineering advances which have been made in the control of the emission of effluents by means of filters and precipitators of a variety of types. Though this work is interesting and very important it has to do with the control of known entities, whereas the vital issue in air pollution has been the discovery of the nature of what was in the atmosphere, so that in turn the source of such entities could be found. The story is intimately bound up with the history of the development of industrial hygiene, for very often the discoveries made in industrial hygiene laboratories, in which the health of the workers was the question at issue, were applicable to the work being done in air pollution research, in which the amenities of the city dweller were the basis of action.

1890-1905

In the graphs shown in Fig. 1 and 2 it will be noticed that in the period 1890-1905 there was a steady rise in the number of papers on air pollution produced each year. As Fig. 2 shows, most of the papers were devoted to methods for controlling the emission of smoke. When these contributions are examined it is found that a great many of them were of the nature of " common-sense " attempts to remove smoke, for 1890 was still the era of " common-sense " engineering, during which, in Great Britain, truly amazing achievements in the textile industry, in railroad engineering and in general mechanization had been accomplished by men with no scientific training but abundant mother wit and tremendous perseverance. This was the period in which the phrase " consume its own smoke " was largely used by engineers, administrators and the public, without many of the users really knowing what the term implied. It can be seen that a good many of the control systems which were developed depended on a belief that if smoke were to be heated to a sufficiently high temperature it would be " consumed ". This belief was adhered to in spite of the fact that the chemical section of the report of the London Smoke Prevention Exhibition of 1883

(Frankland, 1883) had made a most exhaustive analysis of the nature of combustion and shown that not only a high temperature was needed but also a sufficient supply of oxygen. However, engineers in general were not in close contact with chemists, and there is an obvious time-lag between the thorough analysis of the chemistry of combustion and the beginnings of a sound application of this knowledge to designs for almost complete combustion.

In the period before 1900, however, there were several noteworthy publications. Among these were five papers on damage to plants from sulfur dioxide, in one of which (Watson, 1899) workers in Kew (near London) remarked that damage can be prevented by deliberately retarding plant growth (causing the plant to rest) during a fumigation period—a fact which has recently been rediscovered in California. In another article (Ost & Wehmer, 1899) it is shown that the brown or red spots on the leaves of plants consist of air-filled collapsed cells—a finding which has been recently demonstrated in the case of Los Angeles smog by Bobrov (1952).

At this time also the original article in which Ringelmann (1898) described his chart for estimating smoke blackness was published, and the first paper appeared describing a light-absorption method for estimating quantities of pollutant in fine particulate form (Fritzsche, 1898). Witz, writing in 1885, suggested that the increased sulfur dioxide in the air over Paris was responsible for a reduction of the ozone content of that air during a period of ten years (a theory which has again been advanced in Los Angeles, where the lack of sulfur dioxide has been thought to allow the building up of a high concentration of ozone and other oxidants). In addition, two papers described techniques for obtaining deposits of smoke on a revolving paper, in which it looks as if the principle of the thermal precipitator were being invoked (*Stationary Engr.*, 1892; Thomson, 1892).

Thus in this period, although there was a great deal of activity of a non-scientific type, including a proposal to suck fog through pipes laid in the streets and discharge it through large chimneys—strongly resembling a similarly foolish proposal made in Los Angeles in 1945—a great many fundamental principles concerning the origin, nature and dispersal of air pollutants had been stated with a certain amount of precision.

1905-1915

During this period interest in the effects of air pollution greatly increased and the graphs show that research activity caught up considerably on technical activity. A good deal of the medical interest was of a clinical and epidemiological nature, and, as can be expected, there were papers which contradicted one another, for opinions based on clinical impressions cannot be very reliable. The general level of the medical discussion was that there were undesirable clinical effects from smoke, sulfur dioxide and other pollutants, but there was no consensus of opinion that general city pollution was fatally toxic. However, there was one paper (*Sci. Amer.*, 1914 b) which claimed that cancer had a higher incidence in places where coal is used. During this period also a good many of the city surveys were undertaken in the USA in an attempt to find a basis on which to plan a smoke control system, and a general survey of smokiness was begun in British cities.

Activities in Germany were mostly in the direction of (*a*) assessing the damage done to vegetation by sulfur dioxide, for the purpose of establishing the compensation which was to be paid under the German laws governing damage done by effluents; and (*b*) studying the dispersal of gases from factory stacks, though it would seem from a review of the titles of the papers that much of the work was done at the level of industrial investigation and was not carried to the stage of fundamental research. Consideration of possible harm to health through air pollution was apparently not given much attention in Europe.

1915-1925

During this period war, and the readjustment after it, caused a marked drop in the level of activity connected with air pollution. Almost nothing was written on the subject of the effects of air pollution, and the few papers on methods of determination of pollution are all of American origin and mostly of a review nature.

In the section on control methods there is not much of interest, but three points may be noted: (*a*) it is clear that engineering thinking had realized that the prevention of smoke emission from boilers is very largely a matter of correct design of the furnace and not of adding " smoke consumers " to badly functioning furnaces (Buell, 1923); (*b*) references to the use of powdered fuel and to the fly ash which it produces began to accumulate; and (*c*) in 1915 the report of the Selby Smelter Commission was published (Holmes, Franklin & Gould, 1915), and this is important because it is one of the earlier instances in which scientific assistance to a company whose effluent was a serious danger to the surrounding country resulted in the recovery of sulfur as a by-product and in the installation of continuous recording instruments to keep a constant check upon the sulfur dioxide concentration in the air round the smelter. Selby is also the owner of the highest stack in the world (650 feet, or about 200 m), as the result of an appreciation of the effect of high stacks in reducing the concentration of pollution.

1926-1935

In this period the most dramatic event was the Meuse Valley disaster, in which over sixty persons were killed by polluted air (Firket, 1931; Haldane, 1931; van Leeuwen, 1931), and this triggered off activity throughout the world. French works begin to appear in the bibliographies, and the dangers of air pollution were discussed in the French Chamber of Deputies. In the French papers published at that time there was considerable interest in the work being done in Great Britain and also in the USA, but there were almost no references to French writers in the past (with the exception of Bordas (Bordas, 1926; de Boissezon, 1927), who had worked in Great Britain, the USA and Germany), so that it appears that very little work had been done in France before that time.

The German bibliography (Schwartz et al., 1955) lists for this period ten articles on the effects of air pollution of which five are on damage to human health, whereas in the period before 1925 almost all the papers listed deal with the effects of air pollution on vegetation.

Thus the medical scientists, who had been to the fore in the years before 1890, but whose speculations at that time were based more on conjecture than on factual knowledge as to the specific damage done by air pollution, were once again able to take a lead in directing interest to a subject worthy of very active physiological and clinical study.

Work on methods of measurement of pollutants is represented by the statement of the British Department of Scientific and Industrial Research that a close relationship had been established between the solid content of a smoke-stack plume and the optical density of that plume, as measured by the Ringelmann chart or other instrument. This was a considerable advance, for it gave the chart measurements an additional authority which was much needed. In the USA, a paper by Drinker & Hazard (1934) described an apparatus for the measurement of dust, consisting of a travelling transparent tape on to which the dust was blown through a nozzle, thus giving a permanent record. It is probable that the immense labour involved in examining such a tape is the main reason why this instrument has not been more used. Both in the USA and in Great Britain instruments were being developed for the measurement of smoke inside boiler flues, as a means of assisting in the control of stoking. In these two countries and in Germany attention began to be drawn to the dangers of the exhaust from internal combustion engines (Florentin, 1928; Sayers et al., 1929). At that time the emphasis was on carbon monoxide, for it was shown that concentrations of this gas in the streets of the larger cities often came close to the threshold of tolerance. In addition, a couple of papers appeared pointing out that air pollution contributed considerably to the hazard of flying (Aldworth, 1930; Connolly, 1934). This is an important point and it is surprising that so little has been done by the aeronautical technicians of the world to draw more attention to this aspect of air pollution.

This ten-year period started well with research work in the USA, but activity in Great Britain and other countries of Europe was reduced by unemployment conditions. An economic crash in the USA in 1928 had an adverse effect on research, and it is likely that the Meuse Valley incident would have produced a much greater development of interest if economic conditions had been more favourable.

1936-1945

At the beginning of the period 1936-45 there was a noticeable increase in research and technical activity. Interest grew in the nuisance value of fly ash from boilers, and the " dust bowl " dust storms of central USA aroused considerable interest in silicious and other dusts as a public health hazard. The US Public Health Service undertook to make a nationwide survey of the extent of city air pollution and started a survey of the smoke, soot and sulfur dioxide concentrations in the air of fourteen American cities (Ives et al., 1936). The aftermath of the Meuse Valley disaster (six years before) produced at least four papers discussing the possible causes of the deaths which occurred, and the blame was laid on sulfur dioxide by some authorities and on fluorides by others.

A monograph appeared on fluorine intoxication (Roholm, 1937), indicating the industrial activities which produced fluoride effluents and describing the hazards to vegetation, animals and man. In 1937, two articles appeared, one emanating from France, on the production of aldehydes by internal combustion engines (Maillard, 1937; Pardoe, 1937), in which it was shown that the aldehyde concentration in the exhaust was forty times greater during the deceleration period (with throttle closed) than it was during acceleration or normal running. In the following year four more articles were published on the same subject (Grunder & Rich, 1938; Maurin & Kling, 1938; Pardoe, 1938; Roberts, 1938), and one of them indicated that similar effluents could be expected from oil-burning furnaces if the air supply was too great and incomplete combustion took place. Thus at this early date information was being collected which should have been of considerable value to the investigators in Los Angeles ten years later. At this time (1937) the Department of Scientific and Industrial Research reported that the smokiness of many cities in Great Britain had been markedly reduced by the depression, but the war which followed made conditions very much worse than they had been before. A Japanese paper described a photo-cell method of measuring the optical density of filter papers containing particulate air pollutants (Kashiwai, 1938). This of course was not a new process except for the use of the comparatively new photo-cell, and it is doubtful if the Japanese publication advanced scientific methods appreciably. The investigation of the massive sulfur dioxide emissions from the stacks of the Trail smelters in Canada (which produced considerable damage across the border in the USA) had begun in 1929, but a preliminary report was presented by the international commission only in 1937, and it was not until 1941 that a final decision was arrived at (Dean & Swain, 1944). This scientific study of industrial pollution is historically important because it was the first case in which meteorological control methods were specifically applied to industrial pollutants, the industry undertaking to shut down certain processes whenever the meteorological observations taken in the neighbourhood of the smelters showed that conditions were right for the production of atmospheric stratification and a resultant fumigation in the valley running south from the smelter stacks.

The year 1942 is interesting for the appearance of a paper on the toxic effects of pollutants (Easton, 1942), in which it was suggested that the presence of two pollutants, both in concentrations below the accepted tolerance threshold (or M.A.C.), could produce toxic effects because the effects were additive. This is not the first reference to synergistic effects with regard to air pollutants but it is certainly an early one.

In 1943, Great Britain, in the midst of a total war, was still able to hold a conference to consider the claims of clean air in the reconstruction of British cities when the war should come to an end (National Smoke Abatement Society, 1943). In 1944 came increased interest in meteorological factors affecting pollution concentration (Fletcher & Smith, 1944), and the suggestion that the US Weather Bureau statistics would be of assistance to control authorities in the cities of the USA (Fletcher, 1945). In 1945 the Los Angeles Air Pollution Control Office, which had been working on smog control since 1942, issued a paper on the various sources of sulfur dioxide, which was then thought to be the main cause of eye irritation (Swartout & Deutch, 1945). At the same time the report on the Leicester Survey in Great Britain, whose publication had been delayed during the war, appeared, drawing attention to the important part played by turbulence in the dispersion of pollutants from the air above a city (Great Britain, Department of Scientific and Industrial Research, 1945).

In this period, according to the information available, German interest grew considerably, and twenty-eight papers are known of which contributed to knowledge of such matters as dispersion of dust from stacks (Löbner, 1937a), meteorological factors affecting dispersal of pollutants (Lettau & Schwerdtfeger, 1937; Roetschke, 1937), the measurement of sulfur dioxide (Müller, 1941; Schepp & Frömme, 1938), carbon monoxide (Bayer, 1938; Wirth & Muntsch, 1940), and dust concentrations (Löbner, 1937b).

There is not much information on French activity except for a couple of papers on aldehydes from internal combustion engine exhausts, an article on smoke in Paris stating that 90 % of coal burned in Paris is used in domestic grates (Kling, 1938), and some medical discussions of the Meuse Valley disaster.

1946-1957

In this period research developed very rapidly, as is evidenced by the number of papers published and the variety of subjects studied. There was a sudden development of activity immediately the war ended, but it looks as if for some years the output of scientific and technical articles remained steady at about 150 per year. The number of papers is far too great for it to be possible to review all of them, but mention will be made of a few of the most interesting examples.

The 1948 Donora disaster was a historic event and stimulated even more research than did the Meuse Valley disaster, but in addition it caused scientific interest in air pollution to develop in South Africa and Australia and in other young countries that were just arriving at the point where city populations had reached the level at which danger from pollution arises. The Donora disaster also stimulated a considerable amount of physiological and epidemiological research into the nature of the reaction of the human system to air pollutants, and work on this subject is progressing in many American universities and research institutes, and in a number of laboratories in Canada, Great Britain and the continent of Europe. The past ten years has seen the development of several instruments for the measurement of pollutants, such as the American Iron and Steel Institute smoke sampler, the Gast air sampler, the Hi-vol air sampler, the Thomas autometer for gaseous pollutants, the titrilog, the Salzman reagent in a modified recording colorimeter for nitrogen dioxide, the infra-red carbon monoxide recorder, the mass spectrometer for the analysis of hydrocarbons, the rubber-cracking technique for the estimation of ozone, and the colorimeter for oxidant determination. The work of this last period, however, deserves a section to itself.

The Modern Era in Air Pollution Study and Control

The year 1945 can be taken as the start of the modern era in the study of air pollution. First, it is the year in which general activity increased so rapidly that the number of publications appearing in 1947 was four times greater than in 1944. Secondly, by 1945 the Los Angeles smog, which first attracted attention about 1940, had developed to serious proportions and the Control Office, which had been established in 1945, was reorganized as a County Control in an endeavour to meet the crisis. Thirdly, in 1948 the Donora disaster roused the US Public Health Service into intensive activity and convinced a very large percentage of the scientists in the USA that even if clear evidence could not be presented that pollutants such as sulfur dioxide in low concentrations produced physical damage to the human body, there was at last proof that under some circumstances air pollution in the streets of a town could be accompanied by a death rate high enough to frighten public health officials. The London disaster of 1952 confirmed this fact, but even before that date the whole tenor of air pollution research in the USA, Great Britain and on the continent of Europe had changed. Research activity was heightened, money grants for study and city control schemes were enormously increased, and a conviction grew that the pollutants of the air in the modern era were not only smoke, sulfur dioxide, ash and the chemically known gases which are effluents from industries, but a number of substances which had never before been suspected to exist in the normal city atmosphere (and which possibly had not existed there before the year 1900). These substances, whose presence was first detected in Los Angeles, were thought to exist at low concentration in all modern cities, and the characteristic of air pollution study in the present era is a concerted effort to discover what the substances are, for until they are identified it will be difficult, even impossible, to remove them. In addition, in about 1946 the realization had come that air pollution is an area problem, not a city problem. The control techniques therefore began to change from city units to county units in the USA, and the planners in Great Britain turned to national legislation as the only way of dealing with the situation.

Thus it may be said that the years 1945 to perhaps 1953 were a time of mental reassessment of the whole problem of air pollution, while the years since then have been a period during which a move has been made to marshal the forces of all the branches of science into a concerted attack upon what is now realized to be a problem concerned with highly complex organic chemical reactions and a multiplicity of physiological reactions produced by the chemical by-products.

It has been realized that there are a number of fields which need to be explored, and work has been developing very intensively under the following headings:

(1) The development of instrumentation for the continuous recording of the many parameters (ozone, aldehydes, oxides of nitrogen, oxides of sulfur, oxides of carbon, hydrocarbons, etc.) which need to be studied.

(2) Studies of the chemical composition of the atmospheres of large cities with a view to finding out which gaseous compounds are common to all cities, or at least to many cities, and which are peculiar to some cities. In this work the mass spectrometer and the infra-red spectrometer have been the most valuable tools, the one assisting the other.

(3) Attempts to unravel the complex chemical reactions which apparently give rise to the type of pollutant that causes the eye-irritating smog in Los Angeles. This problem is being approached both by the synthetic method, in which compounds are produced and their biological effects are tested, and by the analytical method, in which attempts are made to separate out of normal air some component which will produce specific biological effects.

(4) Toxicological studies of the effects of various materials, particularly ozone, on animals, and population studies in which attempts are made to correlate general population symptoms with the rise and fall of various factors in city air. Here of course the industrial hygiene experts are coming

into even closer contact and co-operation with the workers on air pollution than they did before, and it is only too obvious that air pollution studies are an extension of industrial hygiene.

(5) Meteorological studies to develop a sound knowledge of the nature of air movement in the first few hundred feet above a city, the mechanism by which pollutants are dispersed above a city and the sort of measurement which will most reliably indicate what degree of dispersal rate is in process at the time of the measurement.

(6) Study of damage to vegetation as a means both of learning how the damage takes place and of being able to use specific plants as a form of instrument to indicate the degree of pollution which is present at any time.

(7) Development of methods for the control of effluent from all types of industry and all processes which may produce effluent, including domestic burning of coal, and burning of petrol in private automobiles.

The first attempt to marshal scientific manpower in the USA was the government-sponsored technical conference on air pollution which was held in Washington in 1950 (McCabe, 1952). Another is represented by the national air pollution conferences which were sponsored in California by the Stanford Research Institute in co-operation with the California Institute of Technology, the University of California, the University of Southern California, the Air Pollution Control Association and the Air Pollution Foundation (*National air pollution symposia*, 1949, 1952, 1955).

The most important move was the decision by the US Federal Government in 1955 to vote five million dollars annually for the promotion of research on air pollution, and its administrative action in establishing a research division on air pollution at the Robert Taft Sanitary Engineering Center, Cincinnati. This move has produced a marked increase in research activity throughout the USA and has enabled the Robert Taft Center to act as a clearing-house for ideas about research projects and for the pooling of the results of research. The British authorities have been strong in urging that there should be a pooling of effort and of results, for they have realized, as have all workers in this field, that the development and the discovery of the complexities of air pollution are just an indication of a new factor in society which has developed as a result of the technical advance of mankind. In Great Britain, because of its small size and high population density, research has been fairly well co-ordinated for some considerable time, so that the move to establish close relations with the American workers is a natural development.

Another subject of study which has been considered in this reassessment of the problem is that of the spread of radioactivity as a result of the explosion of nuclear weapons. This is a specialist department of air pollution research which is at present being handled by departments of the atomic energy organizations of the various countries, but it is very apparent that with the development of nuclear energy for industrial purposes the whole subject of the dispersal, monitoring and control of radioactive effluents will have to become the province of scientists concerned with air pollution in general. It is not surprising, therefore, that more and more papers concerned with this aspect of air pollution are being published, and that in the co-ordination of research on air pollution, radioactive pollutants are being given serious thought.

The following references are indicative of the type of work which is being done in these different directions: Beckman & Co., 1955; Billings, Small & Silverman, 1955; Bobrov, 1955; Cadle & Magill, 1951; Cleeves, 1953; Eichfelder, 1955; Eisenbud et al., 1949; Gucker, Pickard & O'Konski, 1947; Haagen-Smit, 1952; Hewson, 1951; Hueper, 1954; LaBelle et al., 1955; Littman, 1956; McCabe, 1956; Magill et al., 1950; Noble, 1955; Patton & Lewis, 1955; Shepherd et al., 1951; Sherlock & Stalker, 1941; Sinclair & LaMer, 1949; Sutton, 1949; Sutton, 1950; Tebbens, Thomas & Mukai, 1956; Thomas & Ivie, 1946.

Comments on the History of Air Pollution

At the end of this review of the history of air pollution it might be of value to comment on the factors of importance which seem to emerge from the study.

(1) The first comment is that air pollution is a subject which seemed to be rather simple at first sight, but which has been found to be more and more complex as it has been explored. Not only is it now known that the oxidant type of pollutant which is associated with what might be called a " hydrocarbon economy " is extremely complex in nature, but it is beginning to be suspected that the reducing type of pollution—referred to in general as sulfur dioxide—is also very complex, a supposition which could account for the fact that concentrations which used to be thought of as quite harmless have been shown to cause considerable injury to health.

(2) The result of this complexity is that though it is fairly easy to obtain generalized information about air pollution (the British investigators gained extremely valuable information with apparatus costing not more than £40, or US $112.00) the cost involved in taking the next step inwards towards the heart of the problem rises out of all proportion to the amount of information obtained.

(3) The preceding two statements lead naturally to the observation that a great many of the research projects which have been instituted during the last sixty years have produced inconclusive results (*a*) because the design of the experiments was inadequate for the problem, or (*b*) because the experimental and organizational structure was deficient, owing to the fact that the funds available were quite insufficient for the task in hand.

(4) It is noticeable in studying the history of air pollution that there has been a tremendous amount of repetition of experiments, discoveries and observations. This means that the waste of effort on the research side has been enormous, but it is not at all certain that effort has been wasted on the social side. It is quite evident that the implications of air pollution to society in the spheres of health, amenity, comfort, and straight cost have never been clear to the community in the past sixty years, in spite of the efforts of control associations. Thus the continual production of scientific review articles giving a general idea of the issues involved in air pollution has rendered a very valuable service in forming in the public mind a desire to deal with the problem, a realization that it can be dealt with, and a willingness to supply the funds which are necessary to deal with it. Notwithstanding this consoling thought, however, in many cases the pooling of money and of workers could have produced experimental projects which would have reached a conclusive end-point; instead one is left with tantalizing suggestions of what might have been found if only something had been done better.

(5) The next comment is that it is very necessary for workers on the same subject in different cities, let alone in different countries, to have a common basis of understanding. In the past, varying tests for the same factor have been used, and it has been impossible to compare the work of one research group with that of another because the units were not the same, the techniques were different and conversion factors could not be applied. Thus the result obtained in one area could not help to achieve an advance in another area. This has been appreciated by most students of the subject, and there is no lack of willingness to pool results and make the pooling as simple as possible.

In this connexion the report of the Air Pollution Planning Seminar which was held in Cincinnati in December 1956 should be cited. A great many of the issues which have been raised above were considered in that seminar and the 176 pages of the report are full of quite fascinating and most encouraging reading (US Department of Health, Education, and Welfare, 1956).

(6) It is no longer possible to think that any country, which is not still to be described as having a purely agricultural economy, can afford to be ignorant about air pollution. Such pollution as is observable in the cities of the world may be slight where populations are small and techniques are not highly developed, but there is no hope that the concentration levels will remain low. As industrial development takes place, pollution will have to be controlled, and a country can ill afford to be without the services of men trained in the investigation and the control of pollution in the air.

(7) Finally, it has been amply proved that air pollution cannot effectively be controlled by placing the control in the hands of the public health organizations of a city, a state or a country. The requirements are diverse

and complex and a team of trained minds is necessary. Medical, physical, chemical and engineering abilities are needed and must be supplied if the job is to be done effectively.

Thus well planned effort is required in every country of the world (the level of the effort adjusted to the level of social development), and there is a great need for these national efforts to have a considerable measure of coherence, so that advance on one front can immediately be used to make easier the work on all other fronts. The beginnings of such an approach are very evident, and it is worthy of much time and thought to make this approach world wide.

REFERENCES

Aldworth, R. (1930) *Hazards in airports due to smoke.* In: *Proceedings of the Smoke Prevention Association of America ; 24th Annual Convention,* Pittsburgh, p. 8

Bayer, F. (1938) *Gasanalyse,* Stuttgart, p. 127

Beckman & Co. (1955) *Chem. Week,* **77**, 70

Beilby, G. (1899) *J. Soc. chem. Ind. (Lond.),* **18**, 643

Billings, C. E., Small, W. D., & Silverman, L. (1955) *J. Air Poll. Contr. Ass. Amer.,* **5**, 159

Bobrov, R. (1952) *The anatomical effects of air pollution on plants.* In: *Proceedings of the second national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 129

Bobrov, R. (1955) *Amer. J. Bot.,* **42**, 467

Boissezon, P. de (1927) *Ann. Hyg. publ. (Paris),* **5**, 1

Bordas, F. (1926) *Ann. Hyg. publ. (Paris),* **4**, 701

Buell, D. C. (1923) *Making friends.* In: *Proceedings of the Smoke Prevention Association of America , 17th Annual Convention,* Pittsburgh, p. 44

Builder (Lond.), 1899, **77**, 143

Cadle, R. D. & Magill, P. L. (1951) *A.M.A. Arch. industr. Hyg.,* **4**, 74

California State Department of Public Health (1955) *Clean air for California,* San Francisco

Carpenter, A. (1880) *J. roy. Soc. Arts,* **29**, 48

Cleeves, J. C. (1953) *J. Inst. Fuel,* **25**, 323

Connolly, J. I. (1934) *Amer. J. publ. Hlth,* **24**, 260

Davenport, S. J. & Morgis, G. G. (1954) *U.S. Bur. Mines Bull.,* No. 537

Dean, R. S. & Swain, R. E. (1944) *U.S. Bur. Mines Bull.,* No. 453, p. 304

Drinker, P. & Hazard, G. W. (1934) *J. industr. Hyg.,* **16**, 192

Easton, W. H. (1942) *Industr. Med. Surg.,* **11**, 466

Eichfelder, K. (1955) *Erzbergbau u. Metallhüttenwesen,* **8**, 111

Eisenbud, M. et al. (1949) *J. industr. Hyg.,* **31**, 282

Firket, M. (1931) *Bull. Acad. roy. Méd. Belg.,* **11**, 683

Fletcher, R. D. (1945) *Meteorology and smoke abatement.* In: *Proceedings of the Smoke Prevention Association of America ; 39th Annual Conference,* Pittsburgh, p. 31

Fletcher, R. D. & Smith, D. E. (1944) *Meteorological factors affecting smoke pollution.* In: *Proceedings of the Smoke Prevention Association of America ; 38th Annual Conference,* Pittsburgh, p. 123

Florentin, D. (1928) *C. R. Acad. Sci. (Paris),* **185**, 1538

Frankland, E. (1883) *Nature (Lond.),* **27**, 407

Fritzsche, P. (1898) *Z. anal. Chem.,* **37**, 82

Great Britain, Department of Scientific and Industrial Research (1945) *Atmospheric pollution in Leicester ; a scientific study*, London

Great Britain, Department of Scientific and Industrial Research (1956) *Fuel Research Pamphlet [s]*, London (NL 110, NL 113, NL 118 - Cyclostyled)

Grunder, L. J. & Rich, P. C. (1938) *Motor coach gassing ; report*, Los Angeles, Los Angeles Railway Corporation and Richfield Oil Corporation

Gucker, F. T., Pickard, H. B. & O'Konski, C. T. (1947) *J. Amer. Chem. Soc.*, **69**, 429

Haagen-Smit, A. J. (1952) *Industr. Engng Chem.*, **44**, 1342

Hahr, H. (1928) *Gesundheitsing.*, **51**, 497

Haldane, J. S. (1931) *Brit. med. J.*, **1**, 366

Heller, W. A. (1954) *Gesundheitsing.*, **75**, Heft 23-24

Hewson, E. W. (1951) *Meteorol. Monthly*, **1**, 5

Holmes, J. A., Franklin, E. C. & Gould, R. A. (1915) *U.S. Bur. Mines Bull.*, No. 98, p. 528

Hueper, W. C. (1954) *Industr. Med. Surg.*, **23**, 463

Ives, J. E. et al. (1936) *Publ. Hlth Bull. (Wash.)*, No. 224

J. Franklin Inst., 1897, **143**, 393

Kashiwai, T. (1938) *J. med. Ass. Formosa*, **37**, 1753

Kling, A. (1938) *Rev. Hyg. Méd. prév.*, **60**, 444

Koritnig, O. T. (1943) *Chem. Zbl.*, **1**, 1625

LaBelle, C. et al. (1955) *A. M. A. Arch. industr. Hlth*, **11**, 297

League Amer. Munic. Bull., 1907, **7**, 7

Leeuwen, W. S. van (1931) *Münch. med. Wschr.*, **78**, 49

Lettau, H. & Schwerdtfeger, W. (1937) *Meteorol. Z.*, **53**, 44

Littman, F. E. (1956) *J. Air Poll. Contr. Ass. Amer.*, **6**, 171

Löbner, A. (1937a) *Veröff. geophys. Inst. Univ. Lpz.*, **11**, Heft 4

Löbner, A. (1937b) *Gesundheitsing.*, **60**, 97

McCabe, L. C., ed. (1952) *Air pollution. Proceedings of the United States Technical Conference on...*, New York

McCabe, L. C. (1956) *Industr. Engng Chem.*, **48**, 96A

Magill, P. L., Rolston, M., MacLeod, J. A. & Cadle, R. D. (1950) *Analyt. Chem.*, **22**, 1174

Maillard, A. (1937) *Congr. mond. Pétrole*, **3**, 557

Mallette, F., ed. (1955) *Problems and control of air pollution*, New York

Marsh, A. (1947) *Smoke ; the problem of coal and the atmosphere*, London

Maurin, G. & Kling, A. (1938) *Rev. Petrol*, No. 768, p. 49

Meetham, A. R. (1952) *Atmospheric pollution*, London

Müller, P. M. (1941) *Angew. Chem.*, **54**, 537

National air pollution symposia, 1949, 1952, 1955, Los Angeles (Stanford Research Institute)

National Smoke Abatement Society (1943) *Measures for smoke prevention in relation to plans for postwar reconstruction.* In: *Proceedings of London Conference*, London.

Noble, W. (1955) *Agric. & Food Chem.*, **3**, 330

Ost, H. & Wehmer, C. (1899) *Chem. Ind.*, **22**, 233

Pardoe, E. S. (1937) *Bus Transp.*, **16**, 480

Pardoe, E. S. (1938) *Exhaust gas odours*, Chicago (Report No. 101-1 of a Subcommittee of the American Transport Association, Bus Division)

Patton, H. W. & Lewis, J. S. (1955) *Analyt. Chem.*, **27**, 1054

Pearson, J. L., Nonhebel, G. & Ulander, P. (1936) *Fuel Economist*, **12**, 84, 108

Pittsburgh University (1912) *Dept industr. Res. Bull.*, **1**, 16

Raymond, V. (1956) *Rapport du Président de la Commission interministérielle de la Pollution atmosphérique*, Paris

Ringelmann, M. (1898) *Rev. techn.*, **19**, 268

Roberts, E. G. (1938) *Power Plant (Engng)*, **42**, 111

Roetschke, M. (1937) *Veröff. geophys. Inst. Univ. Lpz.*, **11**, Heft 1

Roholm, K. (1937) *Fluorine intoxication*, London
Sayers, R. R. et al. (1929) *Publ. Hlth Bull. (Wash.)*, No. 186
Schepp, R. & Frömme (1938) *Papier-Fabrikant*, **36**, 178
Schwartz, K. et al. (1955) *Mitt. Verein Grosskesselbesitzer*, **34-35**, 459
Sci. Amer., 1914a, **110**, 43
Sci. Amer., 1914b, **110**, 233
Shepherd, M., Rock, S. M., Howard, R. & Storms, J. (1951) *Analyt. Chem.*, **23**, 1431
Sherlock, H. & Stalker, E. A. (1941) *Univ. Mich. Engng Res. Bull.*, No. 29
Sinclair, D. & LaMer, V. K. (1949) *Chem. Rev.*, **44**, 245
Smoke Prevention Association of America (1922) *Thirty-three city ordinances adopted before 1922.* In: *Manual of smoke and boiler ordinances*, Pittsburgh, p. 96
Stanford Research Institute (1954) *The smog problem in Los Angeles County*, Los Angeles
Stationary Engr, 1892, **11**, 94
Sutton, O. G. (1949) *Atmospheric turbulence*, London
Sutton, O. G. (1950) *J. Meteorol.*, **7**, 307
Swartout, H. O. & Deutch, I. A. (1945) *The smog problem*, Los Angeles, Los Angeles Air Pollution Control Office
Tebbens, B. D., Thomas, J. F. & Mukai, M. (1956) *A.M.A. Arch. industr. Hlth*, **14**, 413
Thomas, M. D. & Ivie, J. O. (1946) *Industr. Engng Chem.*, **18**, 383
Thomson, W. (1892) *J. Soc. chem. Ind. (Lond.)*, **11**, 12
Tucker, R. R. (1945) *Heat. Piping & Air Cond.*, **17**, No. 9, p. 463
United States Department of Health, Education, and Welfare, Public Health Service (1956) *Proceedings of the Air Pollution Research Planning Seminar*, Washington
Watson, W. (1899) *Effects of fog on plants grown in the houses at Kew.* In: *Transactions of the 7th International Congress of Hygiene and Demography*, London, Vol. 5, p. 21
Wirth, M. & Muntsch, O. (1940) *Die Gefahren der Luft und ihre Bekämpfung*, Berlin
Witz, M. (1885) *C. R. Acad. Sci. (Paris)*, **100**, 1385

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THE IDENTIFICATION OF THE AIR POLLUTION PROBLEM

There are many reasons for the interest that has developed during the past ten years in the various aspects of air pollution, but foremost among them are the dramatic incidents in which fatalities have occurred (namely, the disasters in Donora in 1948 and in London in 1952); the Los Angeles air pollution problem, unique in its own right, but with overtones of broad significance; the hazards of radioactive fall-out; public concern with the chronic effects of air pollution; the development of industry and the growth of communities, which produce more air-borne wastes; new kinds of pollution, or well-known contaminants released in new places; widespread rebuilding of blighted city areas; prosperous times which embrace cleanliness as one of the good things of life; better science and engineering and improved control equipment.

Air pollution now interests workers in such diverse professional fields as the physical and biological sciences, engineering, public health and the law. The United States Technical Conference on Air Pollution in 1950 (McCabe, 1952) convened separate sessions on agriculture, analytical methods and properties, equipment, health, instrumentation, legislation, and meteorology. Why is there this great diversity of interest? The answer lies in the very complexity of the problem. In any air pollution situation the first requirement is to know what standards of air cleanliness are to be maintained. Why is it necessary or desirable to maintain these standards—for health reasons or to eliminate a nuisance? If smoke is the problem, is a change in the type of fuel necessary and what will this cost? If hydrocarbons and the oxides of nitrogen are responsible for eye irritation, crop damage and limited visibility, what are the levels and relative concentrations responsible and what steps can be taken to change the underlying chemical reactions? Are grab sample measurements adequate for evaluation or must there be instantaneous and continuous records? Are instruments available or must they be developed and standardized? If automobile and Diesel exhausts prove to be a major cause of the pollution, is auxiliary control equipment available, how much will it cost to install remedial equipment, and does the constituted authority have the power to require it? Foremost, what is meant by air pollution?

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A typical legal definition of air pollution is: The presence in the outdoor atmosphere of substances or contaminants, put there by man, in quantities or concentrations and of a duration as to cause any discomfort to a substantial number of inhabitants of a district or which are injurious to public health, or to human, plant or animal life or property or which interfere with the reasonable, comfortable enjoyment of life and property throughout the state or throughout such territories or areas of the state as shall be affected thereby (Oregon Revised Statutes, 1956). The American Medical Association's Council of Industrial Health has defined air pollution as the excessive concentration of foreign matter in the air which adversely affects the well-being of the individual or causes damage to property (American Medical Association, 1949). Another definition, which includes natural as well as man-made pollution, is as follows: Air pollution is any air-borne substance which is neither water in any of its phases nor a component of dry air.

These definitions have wide latitude of application and may be interpreted to permit either sensory recognition or physical measurement of the pollution. Both methods of identification are widely used and both have their strength and their weaknesses. Usually the first awareness of an air pollution problem is through some effect on the individual. There are strong or unusual odours, visibility is limited, there is eye irritation, the feel of grit under foot, or an acid taste in the mouth. These are highly subjective phenomena but are no less useful in the identification of the air pollution problem because they vary with the individual. In the case of odours, no better means of evaluation is known than the sense of smell of trained observers (McCord, 1952). While sensory perception may provide the first indication of the presence of most contaminants in the atmosphere it is often not possible to detect trace quantities of many air-borne toxic substances or the presence of radioactive matter through the senses. Their identification requires physical measurement.

In Los Angeles a medical and scientific group recommended allowable concentrations for a small number of specific compounds in the community atmosphere (McCabe, 1955). Limits were established for oxides of nitrogen, ozone, sulfur dioxide, and carbon monoxide. These limits in parts per million of air were adopted and have been in force since 1955 (Table 1). The warning level has been reached by ozone and approached by the oxides of nitrogen during heavy smogs.

This is not the first attempt to set limits for air pollution, however. Swain, in 1910, suggested an hourly average of 1 p.p.m. for sulfur dioxide measured at ground level as the maximum permissible discharge for smelters in farming areas (Swain, 1949). About twenty years ago, the Trail smelter set up an operating regime to prevent sulfur dioxide damage to crops in the

TABLE 1
MAXIMUM ALLOWABLE CONCENTRATIONS OF AIR CONTAMINANTS IN LOS ANGELES

Pollutant	Maximum allowable concentration (p.p.m.)		
	Warning	Alert	Danger
Oxides of nitrogen	3.0	5.0	10.0
Ozone	0.5	1.0	1.5
Sulfur dioxide	3.0	5.0	10.0
Carbon monoxide	100.0	200.0	300.0

Columbia River Valley (Dean & Swain, 1944). Plants sensitive to sulfur dioxide can stand prolonged exposures of 0.2-0.3 p.p.m. without any apparent effect. A limit of 0.5 p.p.m. is generally accepted as the maximum allowable sulfur dioxide concentration for sensitive plants during the growing season.

The oldest and best known effort to apply an objective measurement to air pollution was made by Maximilian Ringelmann about the turn of the century (Kudlich, 1955). The chart which bears his name was introduced into the USA in 1908 and is the basis of smoke evaluation in many ordinances in that country. It has served very well as a reference in reading smoke density or training individuals in the observation of black smoke.

These examples represent some of the attempts at an exact definition of air pollution. In fact, the Los Angeles maximum allowable concentrations are admittedly experimental and the Ringelmann chart has its limitations. It may be concluded that like many other facets of air pollution, if definition were easy it would have been agreed upon long before now. As research progresses there will be more exact definition of polluted air, but at present it is defined with considerable variety and some trepidation.

Identification and classification of air pollution are important to understanding and control. Exclusive of pollen, fog and dust, which are of natural origin, about 100 contaminants have been identified. These fall into the relatively small number of types listed in Table 2 (Rupp, 1956).

TABLE 2
AIR CONTAMINANTS

Group	Examples
Solids	Carbon fly ash, ZnO, $PbCl_2$
Sulfur compounds	SO_2, SO_3, H_2S, mercaptans
Organic compounds	Aldehydes, hydrocarbons, tars
Nitrogen compounds	NO, NO_2, NH_3
Oxygen compounds	O_3, CO, CO_2
Halogen compounds	HF, HCl
Radioactive compounds	Radioactive gases, aerosols, etc.

The Committee on Air Pollution of the American Medical Association reported to the Ninth Annual Congress on Industrial Health (American Medical Association, 1949) that since the general interest in the subject of air pollution had been but recently manifested and because too few attempts had been made at a scientific approach to a solution, the findings of the Committee should be limited for the present to the following general statements and observations:

(1) Air pollution means an excessive atmospheric concentration of foreign matter that may adversely affect the well-being of any person or cause damage to property.

(2) Aside from a few specific but dramatic instances there is a lack of scientific evidence that air pollution seriously injures health. However, if the World Health Organization definition of health is accepted—that is, " a state of complete physical, mental, and social well-being and not merely the absence of disease or infirmity "—then air pollution does have public health implications.

(3) Air pollutants are of two types: those which affect health and those which are not known to have such an effect. In our present state of knowledge, most air pollutants are of the latter type.

(4) There are concentrations of air pollutants which, while they may not be health hazards, are nevertheless intolerably annoying and disagreeable. A health hazard need not be demonstrated to establish the need for control of air pollution.

(5) Air pollution should be reduced to that practical level which may be attained by the application of modern science and technology and which strikes a balance between industry's right to reasonable operation and the community's demand for a decent living environment.

(6) Efforts should be made to enlist the co-operation of industries so that studies of their effluent may be undertaken in order to determine acceptable standards for effective limitation of air pollution, and to provide a technical basis for the design or modification of existing and new equipment.

(7) Scientific knowledge is generally available for developing acceptable methods of measuring air pollution. However, there is a need to develop standard procedures for sampling and for assessment of the dusts, aerosols and gases in the atmosphere so that comparisons between different areas may be made.

(8) In many instances sufficient engineering knowledge is available for the design of equipment and processes to treat atmospheric industrial waste and to remove its objectionable constituents.

(9) It is felt that in our present state of knowledge the appraisal of nuisance intensity cannot be made on an absolute basis but that a useful appraisal can be made on the basis of comparison.

(10) The public programme of air pollution should be carried out by personnel with adequate scientific training and experience. Depending on the peculiarities of the local problem, the requirements of personnel may include such skills as those of chemists, engineers, meteorologists and physicians.

(11) What the function of the physician should be in studies of air pollution and control is not yet clearly defined. Despite the fact that air contaminants are often nuisances rather than toxic agents, there is great need for intelligent medical opinion based on research and for dissemination of factual data.

(12) It is the feeling of this Committee that, although certain fundamentals may be inherently common to all air pollution control ordinances, they cannot be made to apply universally, because of differences in municipal development, topography, meteorology and other related factors.

Five years ago a report[1] to the National Research Council Committee on Sanitary Engineering and Environment in the USA commented on the health aspects of air pollution as follows:

> The question of the role of air pollution on the health of a community is so involved that the job of answering it seems almost insurmountable. Even from the standpoint of a relatively severe short-time exposure the answers do not come easily. This is borne out by the fact that the intensive study of the so-called Donora incident failed to reveal conclusive answers to all of the questions which were raised. This can be attributed in a large measure to the fact that investigations of such occurrences take place after they are over and not while they are going on. The general conclusion of the Donora study was that no single polluting agent was present in sufficient quantity to account for the effects on the population but that apparently some combination of contaminants occurred which was responsible. Bearing in mind the difficulties in studying these effects of a severe short-time occurrence it is possible to visualize the problem of evaluating the more nebulous effects of long-time exposure to much smaller amounts of contamination. In the latter case, one has much more opportunity to study the contamination itself and in this phase of the work considerable progress is being made.
>
> An answer to the question of the relation between air pollution and chronic disease must be found because of the many indications that a polluted atmosphere is not conducive to the best of health. The greatest need today is for some type of co-ordination of the many widespread activities which are now going on. There are many outstanding investigations being conducted in specific related fields, but there is no evidence of any fully co-ordinated study of the health of communities having an air pollution problem . . . It is obvious that a determination of the role of atmospheric pollution in affecting the general health of a population is going to require a tremendous amount of painstaking and costly research over a long period of time.

Topographic and meteorological factors are important in any scheme of air pollution control. If cities could be planned with this in mind there would be less objectionable air contamination, but this is rarely possible. Favourable weather conditions may permit the release of air pollutants in one area which cannot be tolerated under less favourable conditions in another place. Topography has an important bearing on whether wastes discharged into the atmosphere will accumulate in objectionable quantities. Valleys crowded with industrial activity are noted for their smogs and were it not for the advantages of transportation and water supply, they would be avoided as industrial sites.

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[1] Unpublished mimeographed report.

Normally, for every 1000 feet (300 m) above the earth the temperature of the air drops about 3.3° F (about 1.8° C). Frequently radiation of heat to outer space from the earth's surface at night upsets this normal lapse rate and a dense cold stratum of air at ground level becomes overlain by lighter, warmer air. When such an inversion occurs the buoyancy of the air is destroyed and pollution concentrates in the denser air at ground level. This is of frequent occurrence in the autumn and winter months and the accumulation of smoke and other contaminants further aggravates the pollution by preventing the sun's rays from warming the ground and adjacent air. Fog is commonly associated with inversions because the temperature of the air at ground level has fallen below the dew point of the water vapour in the air.

In the Los Angeles area the inversion layer is very resistant to turbulence and acts to hold the pollution near the ground. Visibility is greatly restricted at times of low inversion and contaminants are at a maximum. This brief reference to the meteorological phenomena of the Los Angeles area is to point out the need for fundamental data on air pollution control. Detailed meteorological information is not available for many areas but it is essential to a successful air pollution programme.

The economic effects of air pollution are more readily appraised than are the health effects, and the response of animals and plants to certain contaminants can be observed and measured. The deleterious effects of air pollution on fabrics, metals and buildings represent a significant loss of money and resources in every metropolitan area. There is growing concern about the effects of air pollution on the operation of airplanes and ground transportation (Ingram & McCabe, 1957).

Sulfur dioxide, hydrogen fluoride and oxidizing smogs containing ozone and organic peroxides are the most common air contaminants that affect plants (Thomas & Hendricks, 1956). Several other compounds, particularly mercury, ammonia, chlorine, hydrogen sulfide, carbon monoxide and the growth-regulating substances which are used as herbicides, may cause injury to plants when present in the atmosphere. When toxic concentrations are present the more highly sensitive plants may be useful in monitoring the pollution. While plants are good indicators of certain atmospheric impurities there may be similarity between natural diseases and chemically induced symptoms. Therefore the greatest care must be exercised to avoid errors when using them as control or monitoring devices.

During the past ten years fluorosis has developed in cattle in the USA in regions where the air is contamined with fluorides, particularly when in the form of hydrofluoric acid gas (Phillips, 1952). Symptoms first appear in the teeth and then affect milk production. Eventually the cattle lose weight, there is a reduction in the rate of growth, and the bones develop symptoms which are easily diagnosed. It is possible that the size of the particle taken into the system is a factor in the assimilation of fluorine compounds. If feed containing the contaminant is removed when the first symptoms appear in the teeth, the animal will recover and may show no further signs of fluorosis. When ingestion of contaminants has gone far enough to cause overgrowth in bones and knee joints, cattle will not recover from fluorosis.

Fluorides are released into the atmosphere during the reduction of phosphate fertilizers, the reduction of aluminium, the smelting of some ores, and the manufacture of clay products. Where damage to plants and livestock has occurred mechanical equipment has been widely introduced to control the effluent.

In a study made in 1955 it was estimated that there were 9500 communities in the USA which have local air pollution problems, and of these, 2050 have specific legislation concerning air pollution (Stern, 1955). Legislation seeking to abate air pollution may be divided into two general classifications —namely, punitive ordinances which seek to eliminate air pollution by punishing those discharging contaminants into the air, and regulatory ordinances which seek to abate air pollution by preventing the discharge of contaminants into the air.

While air pollution due to smoke is usually the most common pollution problem faced by our cities, gases, fumes, acids and other industrial by-products, as well as automobile exhausts, likewise contribute, and their discharge into the atmosphere is a matter of statutory regulation and control. Many large metropolitan centres have attempted to solve their smoke problems by punitive ordinances, but the concept of air pollution is no longer confined to smoke, and the trend of regulation is to attack the problem at its source, rather than to rely exclusively on the punitive provisions incident to the exercise of police power to abate nuisances.

For example, in the city of Baltimore, smoke ordinances enacted in 1927 were amended in 1956 to regulate and control the pollution of the air within the city limits by the creation of a Division of Smoke Control under the supervision of a Smoke Abatement Engineer (Baltimore City, 1956). The ordinance prohibits the emission of smoke of a density as great or greater than No. 2 on the Ringelmann chart for more than nine minutes in any hour, prohibits the emission of dust or fly ash where the quantity of solids exceeds certain defined limits, and prohibits the sale, use or consumption of solid fuel which does not meet the standard of a smokeless solid fuel. A system of permits is established for the installation of new fuel-burning plants and the reconstruction or remodelling of existing fuel-burning plants, based upon the submission of plans to the Smoke Abatement Engineer. Operation of the fuel-burning equipment in violation of the ordinance may result in the sealing of the equipment or the imposition of fines. Provision is made for appealing to an Advisory Board against decisions, rulings and orders of the Smoke Abatement Engineer.

However, as was discovered by the city, and then by the county, of Los Angeles, air pollution does not respect political boundaries, and the enactment of statutes, rules, regulations and codes by a metropolis are of no effect if the air contaminants of a neighbouring community, borne by the vagaries of the wind, or confined by topographical and meteorological conditions, are spread over an otherwise regulated community. It has become increasingly evident that the control and abatement of air pollution is not a narrow local problem, but one which calls for country-wide, state-wide or interstate regulation and co-operation.

The states of California, Delaware, Florida, Kentucky, Massachusetts, New Jersey, New York, Oregon and Washington have enacted legislation seeking to control air pollution. Regulations promulgated under such legislation place the emphasis on using fuels efficiently so as to avoid the production of smoke and fumes, and on preventing the emission of solids, gases, liquids and vapours by requiring that equipment designed to accomplish this be installed and operated efficiently, rather than on purely punitive measures or the imposition of fines. The elimination of the causes of air contamination rather than the abatement of a nuisance appears to be the goal of present-day legislation.

In the USA the only federal legislation in connexion with air pollution, other than statutes applying peculiarly to the District of Columbia and the city of Washington, is the Air Pollution Control Bill (United States of America, 84th Congress, 1954), which authorizes the Surgeon General of the Public Health Service to conduct research programmes for the elimination and reduction of air pollution in co-operation with federal agencies, state and local governments, air pollution control agencies and other public and private agencies.

REFERENCES

American Medical Association, Council of Industrial Health (1949) *Ninth Annual Congress on Industrial Health,* Chicago, p. 3 (unpublished mimeographed report)

Baltimore City (1956) Ordinance No. 358

Dean, R. S. & Swain, R. E. (1944) *U.S. Bur. Mines Bull.*, No. 453, p. 285

Ingram, W. T. & McCabe, L. C. (1958) *Proc. Amer. Soc. civ. Engrs,* Paper 1543

Kudlich, R. (1955) *U.S. Bur. Mines Inform. Circ.*, No. 7718, p. 304

McCabe, L. C., ed. (1952) *Air pollution. Proceedings of the United States Technical Conference on...,* New York

McCabe, L. C. (1955) *Industr. Engng Chem.*, **47**, 101A

McCord, C. P. (1952) *Properties, physiology, and psychology of odors.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...,* New York, p. 248

Oregon Revised Statutes, 1956, Section 449, 705 et seq.

Phillips, P. H. (1952) *Fluorosis in livestock*. In: McCabe L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 123

Rupp, W. H. (1956) *Air pollution sources and their control*. In: Magill, P. L., Holden, F. R. & Ackley, C., ed. *Air pollution handbook*, New York, Section 1, p. 1

Stern, A. C. (1955) Cited in: *Baylor Law Rev.*, 1956, **8**, 249

Swain, R. E. (1949) *Industr. Engng Chem.*, **41**, 2384

Thomas, M. D. & Hendricks, R. H. (1956) *Effect of air pollution on plants*. In: Magill, P. L., Holden, F. R. & Ackley, C., ed. *Air pollution handbook*, New York, Section 9, p. 1

United States of America, 84th Congress (1954) An Act (Public Law No. 159) to provide research and technical assistance relating to air pollution control. *United States Statutes at Large*, Vol. 69, p. 322

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THE ROLE OF METEOROLOGY IN AIR POLLUTION

Introduction

The atmosphere is the medium in which air pollution is transported away from the source and, for a given source strength, its actions govern the length of time, the frequency and the concentration to which any receptor will be exposed. On the other hand, meteorology plays only a limited part in the control or elimination of air pollution, since, basically, it does not affect the source strength of the pollutants emitted and since the fundamental atmospheric processes which govern the dispersion of material are not now subject to control or modification. There are three ways to correct an air pollution problem: (*a*) eliminate production of the waste; (*b*) install equipment to collect the waste at its source; (*c*) obtain better dispersion of the material in the atmosphere.

Granted that the ultimate elimination of air pollution is through control of the source, the practical problem is one of balancing the economic cost of reduction of pollutants against the amount of reduction that is required to achieve acceptable levels. This amount of reduction for a fixed source strength is directly a function of the meteorological conditions and their variation in time and space over the area of interest.

The basic parameters governing the transport and dilution of contaminants by the atmosphere can be described qualitatively and semi-quantitatively. Such data permit the intercomparison of various geographical locations and/or the determination of the frequency of occurrence of rapid or slow atmospheric diffusion. The predominant characteristic of the atmosphere is its unceasing change. Temperature, wind and precipitation vary widely as a function of latitude, season and surrounding topography; and these general variations are well known and rather well documented.

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Less well known, and certainly less well documented, are other important meteorological parameters which are useful as indices of air pollution—namely, turbulent structure of the wind, low-level temperature and wind gradients. These parameters show large variations both in time and in space. In fact, they are almost the only meteorological factors that may have been materially affected by man, and even then only locally.

Atmospheric pollution, although usually thought of as the product of industrialization, includes not only the substances released by industrial activity but also natural pollution resulting from volcanoes (Wexler, 1951), dust storms (Warn, 1953), ocean spray (Holzworth, 1957), forest fires (Wexler, 1950), natural pollen in spores (Hewson, 1953), etc. Evaluation of the physiological effects of natural contaminants may often be easier than evaluation of the effects of complex industrial contamination. The character and often the source of natural contaminants are usually better known.

In order to appreciate the role of the atmosphere as the diffusing agent, it is necessary to review the physical nature of the processes contributing to the dispersion of material by the atmosphere and the effects of non-meteorological elements, such as topography and geography.

Air Flow

The fundamental parameter in the movement of contaminants by the atmosphere is the wind, its speed and direction, which in turn are interrelated with the vertical and horizontal temperature gradients in the atmosphere, both of large and of small scale. Essentially, the greater the wind speed, the greater is the turbulence and the more rapid and complete is the dispersion of contaminants in the atmosphere. Since temperature gradients, both horizontal and vertical, increase during the winter season, the speed of the wind flow is generally increased. This is particularly true in temperate and polar latitudes but less marked in the tropics, where the seasonal variation is small. However, occasionally in winter prolonged periods of little or no air motion may occur, particularly in the interior of large continental land masses. A study[1] of the frequency of prolonged periods of light air-flow east of the Rocky Mountains in the continental USA shows that such situations occur most often in late spring and early autumn. For a considerable portion of Europe (Jalu, 1955), light winds occur in late autumn and early winter. In addition to the seasonal change, in many locations there is a diurnal change in wind flow which may be even more marked. At most continental locations, it is usual that the night hours are periods of low-level stability. As a result of the effects of negative buoyancy and the increased energy required for vertical motions, pollutants disperse slowly and may be confined in relatively small volumes. The concurrent light, variable wind may even result in a return flow of material across the original source. In contrast, the daytime winds are apt to be more turbulent, of higher speeds, and the vertical motions are enhanced, so that the maximum dilution of material occurs on clear, sunny days.

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[1] "Climatology of stagnating anticyclones east of the Rocky Mountains in the United States for the period 1936-56", by J. Korshover (unpublished MS.).

The local winds may differ markedly from the general air flow that characterizes the region. Along the coasts of continents or of the larger lakes, the temperature differential between the land and water is sufficient to establish local circulations from the sea to the land during the day and from the land towards the sea during the night (Pierson, 1950; Schmidt, 1957). In the temperate latitudes, these sea-breeze régimes are well marked only during the summer, and are masked by the general wind flow during the other seasons. However, in tropical and subtropical areas, they may be the dominant weather pattern and occur with almost clock-like regularity from day to day.

In addition to the sea-breeze régimes of coastal areas, the topography of an area, either that of the pollution source or that of the targets, may be extremely important. It should be noted, however, that a restrictive topography is not a necessary condition for extreme pollution levels if the source strength is sufficiently high. The best examples of this are the recurring London smog episodes, in which topography plays little or no part. However, with the exception of the pollution episodes in London, all major pollution disasters reported have occurred in areas where the air flow was markedly restricted by terrain (Firket, 1936; US Public Health Service, 1949), so that the flow was persistently and continuously channelled to a single direction or confined in a relatively small area. Within a fairly narrow valley, the characteristic daily wind pattern is a flow up the valley and up the slopes in the daytime, owing to solar heating, while just before or just after sunset, the wind reverses, flowing down the slopes and into the valley (Défant, 1951). Contaminants released within the valley may be effectively trapped within a small area for long periods (Hewson & Gill, 1944). In addition, the shielding from the effects of general circulation patterns afforded by the valley walls will result in lower wind speeds in the valley floor than would be the case in comparable level terrain. The local up-slope and down-slope flow in valley regions may occur almost daily at some locations, but in others it is an exceptional phenomenon observed only on occasion. The existence of local flows and their variation in time is one of the major reasons why detailed studies of individual areas are required to delineate comprehensive air pollution régimes (Holland, 1953). These small-scale flows cannot be detected easily or with certainty from existing conventional meteorological networks.

In addition to the variation of wind flow in the horizontal and with time, there is usually a marked difference in wind flow in the vertical. The roughness of the earth's surface, whether natural or man-made, induces mechanical turbulence, which decreases with altitude. Moreover, the heating of the earth by the sun induces thermal turbulence, which is at a maximum near the surface and decreases with altitude. The net result is a decrease, in the vertical, of the gustiness of the wind and a consequent decrease with increasing altitude in the rate of dispersion of contaminants (Magill, Holden & Ackley, 1956).

Turbulence, or eddy motion, is the mechanism by which effective atmospheric diffusion is achieved. Hence, the study of the spectral distribution of eddy energy, now being investigated on a much more intensive basis (Panofsky & McCormick, 1954; Van Der Hoven, 1957), has important implications in the matter of dispersion of air pollution. The total turbulence consists essentially of two components—mechanical and thermal turbulence. Mechanical turbulence is induced by the movement of wind over the aerodynamic rough surface of the earth and is proportional to the roughness of the surface and to the wind speed. Thermal turbulence is solar-induced and is a function of latitude, the radiating surface and the stability of the atmosphere. It is at a maximum during the summer on clear days and at a minimum during the long winter nights. It has not been common to measure directly the effects of solar radiation on the thermal turbulence, but rather to infer the effects from measurements of the vertical temperature gradient. When the vertical temperature gradient of the lower atmosphere is greater than the adiabatic lapse rate, vertical motions are enhanced, and dispersion, particularly in the vertical, is more marked. On the other hand, in a stable atmosphere, when the temperature gradient is isothermal or positive with altitude, considerable energy must be expended in achieving vertical motion. Even with equivalent wind speeds, a stable lower atmosphere usually results in confining pollutants to relatively shallow layers.

A typical daily cycle of temperature gradient over open country on a cloudless day begins with the build-up of an unstable lapse rate, which increases during the daytime owing to strong solar heating and is associated with well-developed turbulence. Just before or shortly after sunset, the air near the ground cools rapidly and a stable lapse rate (temperature increasing with altitude) begins to form. The inversion increases with time in intensity and in depth during the night, reaching a maximum between midnight and the time of minimum surface temperature. During this period, contaminants are effectively trapped within or below the inversion layer with little or no vertical dispersion. It should be noted that contaminants released during stable conditions at the surface are not transported aloft, conversely contaminants released aloft from tall chimneys, etc., are not generally transported to the ground under these conditions (Church, 1949). With the coming of daylight, the ground begins to heat and the inversion is gradually destroyed. This may result in " fumigation " (Hewson & Gill, 1944), the rapid mixing downwards of contaminants which were released aloft during the night. This often leads to high concentrations during the early fore-noon, before the vigorous mixing of fully developed turbulence re-establishes itself to complete the daily cycle. This cycle may be broken or modified by the presence of clouds or precipitation, which serves to inhibit the vigorous convection of the daytime, but also may prevent the formation of strong inversions during the night.

It has been shown that in urban areas where pollution is most likely, the typical lapse rate régime of the open countryside is modified, particularly at night (Duckworth & Sandberg, 1954). Industrial processes, increased heat capacity of urban areas, and roughness of the buildings contribute to thermal and mechanical turbulence, and the enhanced mixing prevents the formation of a surface inversion. This mixed layer, usually 100-500 feet (about 30-150 m) thick, is capped by an inversion whose base in open country would have been at ground level. Such a condition may neutralize the advantage of emission from tall stacks, since the pollutants emitted will be confined within this relatively shallow layer.

In most discussions of air flow, it is assumed for convenience that the wind remains steady in direction and velocity over a considerable period of time and an extensive area. Actually, this is not the case and detailed analyses of wind flow must take these variations into account. Where the wind flow differs from place to place or with time, owing to pressure gradient differences or topography, meteorological trajectory analyses have been extremely useful in air pollution investigations (Neiburger, 1956) in following the action of contaminants released, or in tracing measured contaminants to their probable source. The computation of exact trajectories requires a large number of accurate wind data, but approximate trajectories can often be evaluated from only a few wind observations and still serve useful purposes.

Routine meteorological data are not particularly suitable for short-term time and space studies of air pollution. This is largely due to instrumental and sampling difficulties arising from the use of instruments with different characteristics, dissimilar exposures and differing sampling procedures and periods of observation.

Atmospheric Diffusion

No attempt will be made here to list the various theoretical approaches to the problem of atmospheric diffusion or to list the working formulae which have been developed. Comprehensive treatments of these aspects are discussed elsewhere (Batchelor & Davies, 1956; Magill, Holden & Ackley 1956; Sutton, 1953; US Atomic Energy Commission & US Weather Bureau, 1955), and a review of this problem is currently being undertaken by a working group of the World Meteorological Organization. While the problems are understood in general terms and the formulations are reasonably precise, the mathematical complexities resulting from the variation of the wind and thermal structure of the lower atmosphere have not, as yet, been successfully overcome for the entire range of meteorological conditions. Also, only fragmentary knowledge is available at present on the fundamental characteristics of turbulence, its three-dimensional energy distribution, and time and space variation. In spite of the lack of understanding of turbulence processes, the working formulae permit the calculation of concentrations from individual sources which agree reasonably well with measurement, except for the elevated source during inversion conditions. Proper utilization of these formulae have permitted useful analyses to be made of the pollution levels resulting from a single source. There have been only a few attempts (Frenkel, 1956; Lettau, 1931) to use analytical methods to compute air pollution concentrations from multiple sources such as would exist in large urban areas. This approach has considerable merit, but it entails formidable computation difficulties and, in addition, requires the development of empirical techniques to account for topographical and boundary parameters. In spite of the difficulties, the analytical computation techniques are probably as accurate as the present knowledge of the distribution of sources, their strength and their variability with time, so that extremely precise formulations would not be required to obtain useful results. Iterated analyses of this type would permit the determination of possible recurrence periods of high-concentration pollution, the determination of " chronic " pollution levels, the evaluation of contributions of various source areas under changing meteorological conditions, and the application of a numerical approach to the question of zoning, emission rates and other air pollution abatement measures.

Removal of Contaminants from the Atmosphere

Dispersal of contaminants in the atmosphere does not remove the material but merely dilutes it through an increasing volume. If most of the contaminants were not eventually removed by other processes, the lower layer of the air would have long since become intolerable to man. Contaminants may be effectively removed from the atmosphere by gravitational settling if the particle size is sufficiently large. The smaller particles, which often constitute a large fraction of the material, may be removed by impaction on the surface of the earth, on vegetation or on buildings, etc. Perhaps the most efficient cleansing agent of the atmosphere is precipitation. The larger particles are readily scavenged from the atmosphere by the falling raindrops (Langmuir, 1948). Recently, it has been shown (Greenfield, 1957) how the smaller particles may be accumulated in raindrops and thus removed from the atmosphere. It might be argued that the arid areas would be subject to much higher pollution levels, although observation does not seem to bear this out. However, the truly dry areas are not often regions of extensive industrialization, and Southern California and the west coast of the USA, an area of little precipitation, is an example of the high level of contaminants that can occur in the absence of vigorous air motions and precipitation. Some evidence exists (Gunn & Phillips, 1957) that air pollution may itself slow precipitation-forming processes.

The gaseous contaminants released into the atmosphere will primarily be removed by absorption, particularly in the oceans and in precipitation. There may be other means of chemical combinations and subsequent removal, but these are likely to be much less important.

The character of pollutants may be modified by photo-chemical or chemical reactions or combinations which may accelerate (or retard) the effects of the pollutants on plants, animals, or structures.

Air Pollution and Visibility

Historically, one of the main objections to atmospheric pollution was the undesirable effect of the reduction of visibility by the black, smoky pall that often hung over industrial regions. More recently this reduction of visibility has constituted an important handicap to commercial transportation, particularly aircraft, and occasionally, in extreme conditions, to automobiles. Visibility is not, however, a reliable direct measurement of total air pollution levels, since it is reduced only by the particulate material, such as smoke and fly ash, sufficient to intercept and scatter visible light. Gaseous emissions or radioactive pollutants could conceivably cause more undesirable pollution levels without ever affecting visibility. However, variations in visibility have been useful in evaluating the gross increase in pollution or the effects of pollution control measures. It can be seen from a study of Los Angeles visibility (Neiburger, 1955) that although there was a decrease in average visibility over the past few decades, it was less than should have been expected from the rapid industrial growth of the area during this period. Conversely, the increase in visibility in the St Louis and Pittsburgh areas (Ely, 1955) after the institution of smoke control procedures was a measure of the success of these actions.

Fundamentally, however, the effect of lowered visibility resulting from pollution is important not because of the reduction of seeing distance but because of the reduction in the transmission of solar energy to the ground. During conditions of extreme stagnation, such as the Donora or London smog episodes, the pollution reached such high concentrations that it effected a major decrease in solar radiation, which in turn permitted the lower atmosphere to remain stable for longer periods, so that there was a " feed back " effect.

Meteorological Studies for Air Pollution Investigations

There have been many well-known air pollution studies over urban areas (for example, Leicester, Donora, Los Angeles, London, and Pittsburgh). Some of these studies did not include concurrent meteorological investigations. This has resulted in some ambiguity in the interpretation of the findings, which is best illustrated by a hypothetical example. If concentrations are measured at one or several points for a given length of time, certain variations of the concentration levels will be found. The question is to determine whether these variations are due to the influence of differing sources, to variations of source strength with time, or to variable meteorological conditions. Without a concurrent meteorological programme, only inferences can be drawn as to the weather variations. Since concentrations can change by orders of magnitude in different weather situations, other deductions become less certain if the meteorological parameters are not well known. Because of the large possible variations within and between areas, the requirements for meteorological data depend largely on the topography, the number of sampling stations, the length of record, and the purpose of the study. If mean annual variation of pollution potential is desired, a single representative meteorological station measuring wind, solar radiation, temperature gradient, etc., may suffice to give adequate information. On the other hand, if it is desired to obtain a large number of individual samples, then a much more elaborate meteorological network is required. A recent investigation of air pollution at Louisville (Leavitt, Pooler & Wanta, 1957) describes the problem of obtaining representative meteorological data for use in a multiple sampling programme.

Air Pollution Climatology

The compilation of meteorological data for the purpose of evaluating pollution levels is not a simple task. Those meteorological stations with long periods of record are not usually in optimum locations for air pollution research purposes. In addition, since direct measurements of atmospheric turbulence levels have not been generally made in the past nor are they currently measured, it is necessary to infer from measurements of wind direction, wind speed, and low-level temperature gradient the dilution potential of the atmosphere. If the frequency of wind observations is sufficiently high, say at least one measurement per hour, then the seasonal variations of this quantity and its correlation with precipitation, air pollution levels, etc., are generally satisfactory. The major difficulty with the wind parameter is the representativeness of the location where the wind records were taken. On the other hand, measurements of the vertical thermal structure of the atmosphere have been made on a widespread basis only since the late 1930's, and these early measurements were usually made only once per day. The primary purpose is to measure the temperature structure of the atmosphere to as high a level as possible. In recent years, these observations have been obtained by means of radiosondes, which have a high rate of ascent in the lowest few thousand feet of the atmosphere and are not ideal vehicles for obtaining the detailed structure of the low-level temperature gradient. It has only been lately that radiosonde measurements have been made at more than a few locations as often as four times per day. Thus, the diurnal variation of low-level temperature gradient and its change with season is not truly susceptible to analysis from radiosonde observations. A number of meteorological towers have been erected in connexion with air pollution investigations where continuous measurements of low-level temperature gradient are available. These are relatively few in number and, owing to restrictions of various kinds, are seldom within large urban areas. Even the continuous records of meteorological towers have their drawbacks in that most of these towers are less than 500 feet (about 150 m) high, so that the structure of the atmosphere above this level is largely unknown or must be determined through utilization of captive balloons, a technique almost as unsatisfactory as the conventional radiosonde. The erection in recent years of tall television towers, 1000-2000 feet (300-600 m) high, in many countries has now provided means by which a continuous record of temperature gradient in the most important layer of the atmosphere can be obtained, but the instrumentation and maintenance of such towers involves considerable work and expense.

In spite of these shortcomings, it is possible to draw reasonable inferences from conventional meteorological data as to the frequency of meteorological conditions that would be conducive to high levels of air pollution. This particular phraseology is deliberately used, since there may be areas where the frequency and duration of conditions favourable to high pollution levels are much greater than in other industrialized areas, but where concentration measurements would not show comparable pollution levels, since no source of pollution is present. An evaluation of the meteorology of potential air pollution must ignore present-day pollutant source strength and consider only the meteorological conditions. It is entirely possible that areas now known for the cleanliness of their atmospheres could become areas of pollution with levels much higher than the now smoky areas of the world.

Considerable care must be used in the evaluation of climatological records for interpretation of air pollution. Differing interpretations are required to answer different questions. If, for example, the problem is one of short-term high concentrations of a particular material or persistent low concentrations of the same pollutant (Eisenbud et al., 1949), then the respective time of sampling and the period of averaging of the associated meteorological data should differ considerably. In addition to gross frequency tabulations of the occurrence of unfavourable meteorological conditions, statistical studies of extreme values of the various parameters and, where possible, determination of recurrence frequencies (Itoo et al., 1955) are most valuable. Directly related to health problems is the question of duration periods of extremely unfavourable meteorological conditions. This requires analyses of time series whose various peak values may not be independent. Thus far, theoretical statistics has not solved this problem and an empirical approach for each location must be utilized. Such an approach requires continuous data measurements.

It is possible to obtain useful data from an analysis of the joint frequency distributions of wind speeds, wind direction and stability, wind direction and precipitation, wind direction and speed according to time of day (Wanta & Stern, 1957). Such information may be used to determine the diurnal and seasonal variation of the conditions favourable to high pollution levels and the definition of an area's microclimate in terms of air pollution potential.

As mentioned previously, data will not usually be available for the area of interest or if available for the area will be taken from only a single discrete point within the area. Thus, at least a qualitative determination of the space variation of the pertinent meteorological factors is required. Fortunately, it is often possible to estimate the deviation in wind flow (direction and speed) and the changes in temperature gradient with changing terrain so as to permit the translation of data from one area to another location of interest. More difficult is the question of relating sampling time of the meteorological variables to the air pollution concentrations. The various working formulae for computing diffusion measurements generally refer to sampling the air concentrations measured over a rather short sampling time. Sutton (1953), for example, determined diffusion coefficients appropriate to a three-minute period of sampling. When this sampling time is extended to the order of days, hours, or even months, then these diffusion coefficients are no longer appropriate and adjustments must be made accordingly (Smith, 1955). On the other hand, for these longer periods, simple averages of wind and stability may suffice when direction variations and diurnal changes are taken into account.

Meteorological Parameters

The use of meteorology for predictions of air pollution may range from statistical prediction based on climatological factors; through the intercomparison of two or more areas to determine potential pollution levels; to the prediction of concentrations of short duration from a single source using analytical techniques. There is also the middle range where predictions can be made for extended periods of unfavourable meteorology for broad areas. A recent study carried out for the Tennessee Valley Authority (Kleinsasser & Wanta, 1956) reflects an approach along these lines. However, the prediction of unfavourable meteorology must be followed by control measures if the actual occurrence of high pollution levels is to be avoided. We are just now approaching the point where prediction of area concentrations for industrialized regions is becoming feasible. If a particular contaminant can be identified as becoming harmful when certain levels are reached, then this type of prediction could permit the selective control of specific sources in order to prevent the particular contaminants from reaching undesirable levels in specific zones.

A service forecasting general stagnant conditions which would affect broad areas is now possible from most of the world's meteorological services and even in the absence of control measures may prove useful in identifying areas in which health officials or air pollution control officers will wish to be especially alert and active.

Finally, the prediction of meteorological conditions favourable for potentially high levels of pollution may be used in actually controlling emission. This is normally considered only as a last resort (Magill, Holden & Ackley, 1956) since it may not be economically feasible unless pollution levels rise to critical values. Actually, meteorological control of all emissions in a large industrial area will probably never be feasible, although the control of emissions from individual sources whose contaminants may be particularly hazardous might be made effective.

Conclusions

In any programme for reducing over-all air pollution levels, meteorology cannot independently provide the ultimate solution. However, it should be an integral part of any air pollution investigation, whether for an individual plant, for an entire industrial region, or for a nation. Meteorology should be considered in the drawing-up of air pollution evaluation programmes. The design of an air-sampling network is intimately connected with the mean air flow and mean stability of the atmosphere. If source regions are well identified, then the meteorology of the area will determine the most economical and efficient location of the sampling network.

Simple consideration of mean air flow and stability will assist in the siting and zoning of urban areas; the development of techniques for computing concentrations from large area sources would be helpful in determining optimum residential areas.

With the increasing urbanization of large areas of the earth, it seems likely that, eventually, pollution from one industrial source (in sufficient concentration to make a significant contribution) will be felt by other areas. This is probably already true in many of the heavily industrialized areas of

Europe and the USA. Since the movement of air is unrestricted by geographical boundaries, the problem is one of international scope. The coordination and interchange of information between all countries working on the air pollution problem is required if progress is to be made in this field. Standardization of instruments and analytical techniques, increasing density of networks, fundamental understanding of the turbulent structure of the atmosphere, and increased utilization of the existing data are indicated for the full realization of the role of meteorology in the understanding and abatement of air pollution.

REFERENCES

Batchelor, G. K. & Davies, R. M., ed. (1956) *Surveys in mechanics*, Cambridge

Church, P. E. (1949) *Industr. Engng Chem.*, **41**, 2753

Défant, F. (1951) *Local winds*. In: American Meteorological Society, *Compendium of meteorology*, Boston, Mass.

Douglas, C. M. K. & Stewart, K. H. (1953) *Meteorol. Mag.*, **82**, 67

Duckworth, F. S. & Sandberg, J. S. (1954) *Bull. Amer. meteorol. Soc.*, **35**, 198

Eisenbud, M. et al. (1949) *J. industr. Hyg.*, **31**, 282

Ely, S. B. (1955) *Report*, Pittsburgh (Bureau of Smoke Prevention, Department of Public Health City of Pittsburgh)

Firket, J. (1936) *Trans. Faraday Soc.*, **32**, 1192

Frenkel, F. N. (1956) *Sci. Monthly (Wash.)*, **82**, 94

Greenfield, S. M. (1957) *J. Meteorol.*, **14**, 115

Gunn, R. & Phillips, B. B. (1957) *J. Meteorol.*, **14**, 272

Hewson, E. W. (1953) *Some aspects of the dispersion of pollens and industrial contaminants in relation to micrometeorology*, Cambridge, Mass., Geophysics Research Division, Air Force Cambridge Research Center (Contract No. AF 19(604)-792)

Hewson, E. W. & Gill, G. C. (1944) *U.S. Bur. Mines Bull.* No. 453, p. 30

Holland, J. Z. (1953) *A micrometeorological survey of the Oak Ridge Area*, Oak Ridge (United States Atomic Energy Commission (ORO-99))

Holzworth, G. C. (1957) *J. Meteorol.*, **16**, 68

Itoo, K. et al. (1955) *Papers in Meteorol. & Geophys.*, **6**, 19

Jalu, R. (1955) *Météorologie*, No 39, 247

Kleinsasser, T. W. & Wanta, R. C. (1956) *A.M.A. Arch. industr. Hlth*, **14**, 307

Langmuir, I. (1948) *J. Meteorol.*, **5**, 175

Leavitt, J. M., Pooler, F. & Wanta, R. C. (1957) *Design and interim meteorological evaluation of a community network for meteorological and air quality measurements*. In: *Proceedings of the Golden Jubilee Air Pollution Control Association Meeting, June, 1957*. St Louis, p. 57

Lettau, H. (1931) *Beitr. Geophys.*, **31**, 387

Magill, P. L., Holden, F. R. & Ackley, C., ed. (1956) *Air pollution handbook*, New York

Neiburger, M. (1955) *Visibility trend in Los Angeles*, Los Angeles (*Air Pollution Foundation Report* No. 11)

Neiburger, M. (1956) *Wind trajectory studies of the movement of polluted air in the Los Angeles basin*, Los Angeles (*Air Pollution Foundation Report* No. 13)

Panofsky, H. A. & McCormick, R. A. (1954) *Quart. J. roy. Meteorol. Soc.*, **80**, 546

Pierson, W. J., jr (1950) *The effects of eddy viscosity, Coriolis deflection and temperature fluctuation of the sea breeze as a function of time and height*, New York (*New York University Meteorological Papers*, Vol. 1, No. 2)

Schmidt, F. H. (1957) *J. Meteorol.*, **4**, 9

Smith, M. E. (1955) *Sampling periods in air pollution evaluations.* In: *Proceedings of the third national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 80

Sutton, O. G. (1953) *Micrometeorology,* New York

United States Atomic Energy Commission & United States Weather Bureau (1955) *Meteorology and atomic energy,* Washington (AECU 3066)

United States Public Health Service (1949) *Publ. Hlth Bull. (Wash.),* No. 306

Van Der Hoven, I. (1957) *J. Meteorol.*, **14**, 160

Wanta, R. C. & Stern, A. C. (1957) *Amer. industr. Hyg. Ass. Quart.*, **18**, 156

Warn, G. F. (1953) *Weatherwise*, **6**, 67

Wexler, H. (1950) *Weatherwise*, **3**, 129

Wexler, H. (1951) *Bull. Amer. meteorol. Soc.*, **32**, 48

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SAMPLING, ANALYSIS AND INSTRUMENTATION IN THE FIELD OF AIR POLLUTION

I. INTRODUCTION

The subject of air pollution has been dealt with repeatedly in the past, from various points of view. However, one has the feeling that in the different meetings the discussions have centred on the more general aspects or on very specialized factors, while rightly stressing the general importance of the problem from the chemical and physical, health, epidemiological, agricultural and zootechnical angles. Such discussions have dealt with the sources of pollution and methods of control and inspection, while the responsibility for pollution has been laid on the ignorance and the lack of interest of town dwellers, the absence of specific legislation and the ineffectiveness of the existing legislation. Finally, some given aspect of a special field has perhaps been dealt with in detail—for example, some specific sampling and analytical technique for one, two or more components of air pollution.

The impression left is one of the initial chaos attendant on all human endeavours, leading to the conviction—at least in the purely technical field with which we are concerned here—that it is necessary to standardize methods and means, as regards sampling and analysis at any rate. Such standardization will yield as clear a picture as possible of the data useful as a basis for general plans for the solution of a problem of such fundamental importance as air pollution.

It is possible to use sampling methods A or B (recommended as the best by different authors) to sample the same impurity X, with considerable differences in efficiency. Moreover, methods C and D (also arbitrarily chosen by investigators as being the best ones) may be used for analysis of the samples, with different limits as regards accuracy, sensitivity and selectivity and, furthermore, with results also affected by the skill, objectivity and working conditions of the operator. While this state of affairs continues, any trustworthy and definitive comparison of results will remain impossible—that is, it will be impossible to deal with a variety of conditions and circumstances in a uniform manner, except within the range of tolerance covering the straying of the results from the true figures which arises from the various causes indicated above.

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For this reason, in the conviction that it is essential to overcome the obstacle of the " subjective " nature of the results, which arises from the disparity of methods and the personal equations of the various workers or experimenters, the author is putting forward in this report, not a definite plan for immediate standardization at this stage, but a preliminary contribution towards selection, from the available material, of those methods which seem most suitable for narrowing the range of " tolerance " already mentioned. It should be pointed out, however, that there are two negative aspects with regard to such a selection: (*a*) the result will, in turn, doubtless show traces of the personal views of those making the selection; and (*b*) the reason why the use of instrumental means in the field of analytical research connected with the problem of air pollution has not yet become general, but has so far been reserved only for the best-equipped research institutes, is essentially a financial one, this question of finance probably being in most cases the main reason for the indifference with which the problem of air pollution is regarded in many countries.

Although the first point may appear relevant, the second—at least as far as we are concerned here—can be regarded as negligible, since we are not considering the drawing-up of a budget of any kind but are only trying to determine the most objective and efficient technical and scientific methods for sampling and analysis. In saying this, we do not wish to disregard the value of the research work carried out by various investigators so far, nor do we forget the future work which will still be necessary. Instrumentation is, in fact, nothing more than the application of new means to old and tried methods, an application which, apart from the benefits it has already shown, will lead to a streamlining of the work as regards both space and time and make it possible to extend the field of application of the methods themselves to situations which could not be investigated by ordinary means.

II. SAMPLING

General

The sampling methods used for the study of air pollution can be classified under three different headings:

(1) Sampling of impurities of every nature, ranging from particulate matter to gases;

(2) Sampling under various environmental conditions, ranging from samples taken from conduits or chimneys to samples taken in the open air;

(3) Sampling methods varying according to the time factor, ranging from intermittent to continuous sampling.

The major difficulties encountered in each of these categories of sampling generally relate to: (*a*) the problem of collecting samples which are genuinely representative of the condition of the mass of air under study; (*b*) the problem of avoiding the introduction of errors arising from the methods used for the separation of the various components of pollution, which is necessary for the analysis of each of these components; (*c*) the problem of preventing any change in the concentration of particulate matter, in suspension as a result of the sampling operations.

Preliminary Considerations and Stages of Sampling : Principles Governing the Avoidance of Sources of Error

Before putting forward a tentative systematic enumeration of sampling devices, we shall discuss a few points of a general nature relating to the principles to be followed to ensure correct sampling, as free as possible from direct or indirect causes of error and capable of giving directly all the necessary information, or making possible its subsequent acquisition.

Statistical studies

Statistical studies are of the utmost importance to establish the basic data (e.g., the size and frequency of sampling) necessary to the drawing-up of a good plan of research. By their use it is also possible to extract the maximum amount of information from the results, thus enhancing the completeness and accuracy of the research. Statistical studies naturally solve such problems in terms of probability—that is, by definition they do not give absolute results but only indicate the percentage probability that a given series of data will be accurate within certain limits.

Size of the samples

The samples should naturally be big enough to make analysis possible. Consequently the persons carrying out the sampling and the analysts should always agree beforehand to make a joint study of the types of sampling devices, the methods of sampling and the size of the samples necessary and adequate for the type of analysis to be carried out.

Changes in the sample during and after sampling

This is one of the fundamental causes of error. In fact, many substances which, when present in low concentrations, can co-exist without reacting with one another and consequently without undergoing any appreciable change, may when sampling has been continued long enough to bring about a high concentration—for example, in order to provide the analyst with an adequate amount of material for analysis—react with the formation of other substances which were perhaps not present either in the waste gases or in the atmosphere. Particulate matter, moreover, may agglomerate or break up, so that the sample collected may show statistical and dimensional properties completely different from what they are in reality. These possibilities should always be borne in mind by the investigator so that he can try to avoid such serious sources of error.

Continuous and intermittent sampling

Automatic, continuous or intermittent recording apparatus is becoming more and more widely used in atmospheric analysis because of the outstanding advantages it offers. Apart from leading to a saving in time and staff, such apparatus is sometimes sufficiently sensitive to detect minute concentrations of impurities that generally escape the notice of even the most experienced workers using the normal analytical means. In some cases, recent research has shown that it is precisely these minute traces of impurities which are the cause of the most unpleasant aspects of air pollution, ranging from damage to plant life to physiological effects in man, such as irritation of the eyes. Many of these continuous recording machines combine the operations of sampling and analysis, so that any differentiation between the two operations for purposes of classification becomes pointless.

Sampling of volatile constituents

The major difficulties involved in the sampling of volatile components of air pollution relate to the efficacy of the methods used, the concentrations which it is necessary to attain, and the subsequent separation of other components in order to analyse the volatile substances. In such cases, the only useful advice of a general nature which can be given is to carry out sampling with large volumes of air so as to have the maximum quantity of the substance available for analysis and to eliminate, as far as possible, the need for subsequent concentration.

Sampling of particulate constituents

It has already been seen how the possible agglomeration or breaking up of particulate matter may lead to errors of evaluation. In order to eliminate these sources of error the sampling should be carried out under conditions which are as isokinetic as possible, above all when the particles are very small.

This signifies that sampling should be performed in such a way that, as far as possible, the gas stream carrying along the particulate matter should undergo no disturbance or change of speed on entering the collecting device.

It is almost impossible, however, to ensure perfectly isokinetic conditions, since the final container will always represent a disturbing factor. Thus sampling takes place practically always under more or less anisokinetic conditions, which may lead to inevitable errors in the determination of both the quantity and the dimensions of the suspended matter. In order to reduce anisokinetic conditions to a minimum, the gas stream should be sampled as far as possible in the same direction and at the same speed as the stream itself, but at least never counter-current.

Furthermore, to avoid reducing the efficiency of sampling, deposits and condensation should not be allowed to form on the walls of the sampling vessel; the collecting surface should therefore be as close as possible to the source of the gas stream. Obviously, a 100 per cent yield of the total pollutant content is not possible, but a fair approximation can be achieved by carrying out selective sampling in relation to the size of the particles.

Sampling of waste gas

Numerous difficulties encountered in this type of sampling arise from the particularly variable characteristics of waste gas—namely, very high temperatures, lack of uniformity in the composition of the gas flow, and differences in speed due to disturbances of all kinds. For all these reasons it is impossible, in the present state of our knowledge, to lay down any general rules, and it is necessary to adopt those methods which are best suited to the special conditions which it is felt will occur in the particular case concerned.

The only general directive which can be given is that the gas stream should be sampled at several points and the largest possible number of samples taken, or that average samples should be taken after a careful study of the conditions of the gas flow, so as to determine as far as possible the variables of the flow and whether these are cyclic or periodic in nature.

Sampling in the open air

In the case of sampling in the open air the main difficulties arise from: (*a*) the high dilution of the pollutants dispersed in the air to be sampled; (*b*) the consequent need to collect large samples (without, however, unduly prolonging the sampling time); and (*c*) the difficulty of sampling under isokinetic conditions, in particular because of the frequently rapid changes in the direction of the wind.

Here, more than in any other case, continuous recording instruments are valuable. Several of them should be placed simultaneously at various points in the area under examination. Synchronous meteorological measurements, again continuously recorded, should also be available for each instrument so as to supply the essential information for identifying at any moment the characteristics of the source of pollution with respect to the wind speed and direction and other basic meteorological data. It is in fact wind and atmospheric stability factors which play the major part in bringing about special conditions of dilution or concentration of pollutants in the air.

Various mathematical formulae have been introduced for defining the theoretical concentrations, detectable at ground level, of gaseous or particulate effluents coming from individual sources (point sources, linear sources, etc.). Such formulae may be of great value, particularly for converting results actually obtained under given meteorological conditions to those which should be obtained under different conditions.

Furthermore, a study of the topographic and orographic features of the area is necessary for the integration of all sampling data, since such features have a considerable influence on the variability of meteorological factors and the limitation of the volumes of air affected by the discharge of pollutants.

Methods for following the movement of air masses

In some cases it may be necessary to analyse the movement of air masses in order to determine the speed of diffusion and the distance at which polluting substances discharged into the air from one or more sources can be detected. In such cases, use may be made of fluorescent substances (zinc and cadmium sulfides, zinc sulfate, etc.), which are discharged into the air by means of a special fog or smoke generator and which are then looked for in samples collected at various distances and at different times in the area under study.

Instruments for Sampling Waste Gases and for Atmospheric Sampling

Devices for general use

Almost all sampling apparatus requires aspirating and measuring instruments, in addition to the collecting device. Almost all, but not all, since occasionally the sample is collected by the displacement of gas or liquid or by the filling of containers which have first been evacuated and which have a known volume. In such cases—nearly all of which are of instantaneous sampling—a measuring device may be unnecessary and the aspirating apparatus can be omitted, provided that the gas to be sampled is under sufficient pressure to displace the liquid or gas filling the collecting vessel. Instantaneous sampling is usually undertaken only in special cases, when it is required to determine the conditions at a given moment, and is employed primarily for sampling waste gases in the immediate neighbourhood of the outlet of conduits or pipes—that is, when sufficient pressure is available for displacement of the contents of the collecting vessels.

However, sampling is generally not instantaneous but more or less prolonged and, as has been said, calls for aspirating and measuring devices as well as, in the case of the direct sampling of a gas stream, probes—that is, sampling devices which can reach any point in the cross-section of a conduit which has been selected for sampling.

Probes

Probes for sampling gas free of particulate matter are generally simpler than those used for gas streams containing solids or liquids in suspension. Such probes are nothing more than tubes suitable for penetrating into the gas stream, and should be constructed of materials which do not react with the substances to be sampled. Since it is not uncommon to encounter special temperature conditions or corrosive matter, probes should be made of stainless steel, or—better still—of glass or quartz.

The length of the probes should be such that they can penetrate to any point of the gas stream to be sampled, since the latter may not be uniform as regards flow, speed or composition. The internal diameter of the probe should be such that it conveys the sample to the receiving vessel at the speed considered most suitable for the particular case. At the same time, the diameter should not be too small, so that the probe does not readily become choked when particulate matter is present and also so as to facilitate cleaning, which should be undertaken as often as possible.

To ensure the isokinetic sampling conditions to which reference has been made, the opening of the probe should face the gas stream to be sampled, and the diameter of the opening should be such that sampling can take place without changing the speed of the effluent.

Meters

Since the precision and accuracy of an analysis largely depend on an exact knowledge of the volume of gas sampled, such devices must be extremely accurate. Meters for the measurement of dry gas are used for preference. They are fitted with manometers and thermometers which indicate, at the point of measurement, the pressure and temperature of the gas stream sampled, so that the volume of gas read off can be converted to standard temperature and pressure, or its weight determined.

Suction devices

No special directions can be given in this connexion, since any suction device may be used which has adequate volumetric capacity under the necessary conditions of reduced pressure. Vacuum pumps driven by electric motors are the most common apparatus used. If there is any danger that the gas to be sampled will ignite, the pumps may be fitted with special anti-combustion motors or, if only very low power is required, they may be worked by hand. Otherwise the gas can be absorbed by using jet pumps employing water or steam, or special pumps driven by a gas motor actuated by liquid carbon dioxide.

Devices for sampling gases and vapours

Absorbers

A gas can be sampled by means of suitable absorption reagents provided it dissolves with sufficient rapidity in these reagents. For this purpose use is made of U-tube absorbers of the type laid down for the determination of sulfur by the American Society for Testing Materials. These absorbers are filled with a certain measured amount of reagent and fitted with a porous glass partition, so that the air or gas led into them passes through the reagent solution in the form of very fine bubbles, thus ensuring intimate contact. Sampling by means of such absorbers is usually carried out at an average rate of about 100-150 litres per hour of gas stream. The absorbers may be arranged in series of two or more elements containing the same reagent solution to ensure more-complete absorption of the impurity in question, or in series of two or more elements containing two or more different reagent solutions so as to absorb different pollutants successively from the same volume of gas or air sampled.

Several years' experience in the Institute of Sanitary Engineering in Milan has shown that units of the type described above can also be usefully fitted up in parallel series. Ten or more units can be used in this way for the simultaneous sampling of several pollutants. The arrangement which at present seems most advisable (according to experience in the Institute) is the installation of parallel pairs of absorbers (mounted in series) corresponding to the number of polluting substances which it is desired to detect.

This arrangement achieves the completeness of absorption considered necessary, and at the same time makes possible the simultaneous sampling of any desired number of pollutants.

By selecting the most suitable absorbent solutions at present available, the gaseous components listed below can be determined under the conditions mentioned in concentrations as low as 0.1 p.p.m. by volume:

oxides of sulfur
oxides of nitrogen
ammonia
hydrogen sulfide
hydrochloric acid
hydrofluoric acid
hydrocyanic acid

Even lower concentrations can be determined by prolonging the sampling time or decreasing, where applicable, the speed of the gas flow through the absorbers.

When the polluting substances collected are present in minute traces and the sensitivity of the analytical method to be employed is very close to the limit of quantity of pollutant which can be collected by absorption, it is possible to improve the degree of absorption by using a more complicated arrangement. A much smaller quantity of reagent is injected as a fine spray into a small bulb, into which the sample of gas containing the contaminant to be absorbed is rapidly and simultaneously injected from another storage vessel by means of an alternate " emptying " and " refilling " cycle. In this way intimate contact is brought about between the reagent and the contaminating substance in the sample, a minute quantity of the component to be determined being concentrated in a very small volume of reagent liquid.

This particular type of sampling by absorption may be employed for the determination of ozone, hydrocarbons or organic solvents and, with suitable auxiliary refrigeration equipment, the samples thus obtained can be used for infra-red and ultraviolet spectrographic analysis.

Adsorbers

The collection of samples by means of adsorption involves considerable difficulty, not so much as regards the apparatus but as regards the need for preliminary investigation to determine what adsorbing medium will give the best results, both quantitatively and qualitatively.

This type of sampling is used especially for ozone and the light hydrocarbons. Adsorption is brought about by aspirating the air or gas to be sampled through adsorption columns containing silica gel, activated charcoal or any other suitable agent. After adsorption, the different pollutants can be extracted from the column in various ways: (*a*) if the adsorption on silica gel has been carried out at the temperature of liquid air (procedure used to detect down to two or three parts per hundred million by volume of ozone), the sample can be extracted by raising the temperature to —120° C; (*b*) the same procedure of raising the temperature serves to separate various fractions of light hydrocarbons from the adsorption column, by taking advantage of the different boiling points of these fractions; or (*c*) a jet of inert gas passing through the adsorption column can act as an eluent and behave in the same way as an elution solvent in liquid chromatography.

Condensers

In this procedure the gas stream sampled is cooled in suitable containers, thus bringing about the condensation of the volatile substances present. As in the case of the absorption devices previously described, these so-called condensation traps can be arranged in parallel pairs and in series at decreasing temperatures. By using various coolants—for example, ice, dry ice, liquid nitrogen, or liquid air—the components can be separated by fractional condensation. However, the quantitative assay of the various pollutants in the sample may be hampered by the formation of crystals, which may first deposit and then volatilize on the less cold parts of the apparatus. This drawback can, however, be reduced to a minimum by dividing off the cooling chamber with suitable porous glass partitions.

Such devices are used in particular for the sampling of odoriferous substances.

Collectors under reduced pressure

For some substances, such as nitric acid and aldehydes, having a high molecular weight, absorption in aqueous solutions is sometimes incomplete. In such cases it is preferable to use bottles of known volume for collecting under a pressure reduced to 200 mm Hg or even less. The absorbent solution chosen is first introduced into the bottle and the pressure is then reduced. Next, the sample is admitted until the internal and external pressures are equal, and the container is shaken continuously so as to ensure maximum absorption.

This method is used especially in the construction of automatic analysers for the oxides of nitrogen. Such analyses range in sensitivity from 0.005 to 5 p.p.m. by volume, and employ a colorimetric reagent, the colour produced being measured photo-electrically.

Samplers for mass spectrometric analysis

Sampling for mass spectrometric analysis can be carried out in various ways, either by compressing the gas sample in a pressure flask so as to concentrate a large quantity of gas in a small volume, or by filling evacuated containers. The sampling technique employed should first be agreed upon with the analyst in charge of the mass spectrometer, since it has a considerable influence on the results obtained.

Plastic containers

Special polyethylene bags, strengthened with at least five layers of plastic material and sealed while hot, are much used for collecting and transporting large volumes of air. These bags have the advantage that they can be used for the successive analyses of small fractions of the sample taken. Moreover, polyethylene is inert with respect to very many substances, among which are sulfur dioxide and formaldehyde. On the other hand, plastic bags are not suitable for collecting and storing aerosol suspensions, because of the possible generation of electrostatic charges as a result of which the aerosols tend to move towards and condense on the walls.

Devices for sampling particulate matter

Sedimentation methods

Sedimentation is a very suitable method for sampling suspensions of pollen, but not dust suspensions, because the particles may clump together or become dispersed over the walls of the sampling device, making it difficult to collect them quantitatively. The sedimentation rate of the smallest particles contained in aerial suspensions may also be considerably lower than that of the larger particles, so that the latter tend to predominate on the sampling slides.

The apparatus for sampling by means of sedimentation consists of a glass surface, measuring about 3 cm × 10 cm, arranged horizontally in a protecting metal frame. The latter consists of two metal discs, about 20 cm in diameter, arranged horizontally and roughly 10 cm apart. The slide is placed in a holder about 3 cm above the lower metal disc. The whole apparatus is mounted on a rod, provided with a support, about 90 cm high, and placed on top of a high building.

The slide becomes coloured and the pollen granules are identified and counted under the microscope, the results being reported as " number of granules per cm^2 of slide collected in twenty-four hours ". The method is simple and economical, but not suitable for quantitative assay of the pollen content of a given volume of air. Particles of homogeneous smoke can, however, be measured by observing their sedimentation rate in a special sedimentation chamber by means of the Tyndall effect produced by a beam of light. In this way it is possible to measure sedimentation rates ranging from 4 cm to 160 cm per hour, corresponding respectively to matter with a density of 1 g/ml and matter with a density of 2 g/ml, such as sulfur.

Sedimentation jars, which are very simple and economical, can also be used for gravimetric determination of the matter deposited in a given period of time, but they give results which are only very approximate and are hardly reliable.

Visual and photometric methods

These are devices for direct observation of the state of dispersion of the particles in the air or in a waste gas, thus eliminating errors arising from the clumping together or breaking up of the particles which may occur when other means are used for collecting them.

The Ringelmann cards are an example of a visual method. They consist of a series of five cards numbered from 1 to 5, divided up into squares formed by black lines on a white ground, the ratio between the areas of the squares on the different cards being 20, 40, 60, 80, and 100 per cent. These cards are placed at the eye-level of the observer, at a distance of about one and a half metres and as near as possible to the source of the smoke. The observer compares the density of the smoke with the intensity of the grey spots which the various Ringelmann cards produce, and describes a source which is practically clear (non-opaque) as No. 0 and a smoke which is 100 per cent black as No. 5. The observations are repeated at intervals of 15-30 seconds and the readings are then related to the theoretical value of the density as determined for Ringelmann card No. 1—that is, a smoke with a 20 per cent density—by means of the formula:

$$\frac{\text{Total of the different numbers of the Ringelmann cards} \times 20}{\text{Number of observations}} = \begin{array}{c}\text{Percentage density}\\ \text{of the smoke}\end{array}$$

This very simple method clearly gives only very approximate results, which may also be completely arbitrary at times, varying according to the position of the observer with respect to the source.

More accurate is the slit ultramicroscope, where the slit is in the form of narrow cell through which is passed the sample at a rate slow enough to ensure that the time of passage is sufficient for purposes of observation. It is usually possible to make about 60 observations per minute and thus obtain a satisfactory statistical average; it is also possible to count particles with diameters of less than 0.1 μ.

Another very sensitive device is a transverse dispersion Tyndallometer. Here the flow of the sample is observed by means of transverse illumination which shows up the particles so that they can be rapidly counted. This can be done directly by an experienced operator or, still better, by electronic counting and automatic recording apparatus. With such apparatus it is possible to count particles of about 0.6 μ in diameter at a speed of 1-1000 units per minute.

Smaller particles can also be observed by using suitable photographic equipment which takes a picture of the sample every two seconds, registering the condensation droplets formed around the particles and thus enabling a subsequent count to be made.

Impingers

This type of sampling consists in the interception of aerosol particles by placing obstacles in the path of the gas stream. When a gas stream is forced to go round an obstacle, the particles in it tend—because of their inertia—to diverge from the direction of the stream and collect on the surface of the obstacle. Of course, since the inertia of a particle is proportional to its mass, the smaller the particles the greater will be their tendency to follow the direction of the gas stream in its flow round the obstacle. The efficiency of this type of sampling is given by the ratio between the number of particles intercepted by the apparatus and the number which would have been intercepted if none of them had been deflected in any way. For maximum efficiency, this method should therefore be reserved for rather large particles, using small samples and a high rate of flow.

There are many types of device employing this principle, ranging from simple microscope slides exposed to gas streams or suspended in the air at right angles to the direction of the wind, to cascade impactors. The latter consist of a series of slides arranged at right angles to one another and it is possible to " select " the particles by gradually reducing the diameter of the opening of the tubes directing the gas stream from one slide to another, at an average rate of flow of approximately 17 m per minute.

Using cascade impactors it is possible to trap particles of submicronic size with the arrangement in series just described, but conveying the gas stream to the last slide at a very high velocity, equal to the speed of sound.

A special arrangement for continuous sampling employs a jet of gas which is made to impinge on a series of slides mounted radially around the circumference of a cylindrical wheel placed in a vacuum chamber, so as to cause the assembly of slides to rotate. More complex apparatus employs a microscope in conjunction with the simple impinger and sometimes also a camera for taking periodic pictures of the slides. In place of the slides tubes containing solvents or inert liquids can be used, in the former case for chemical analysis of the solution obtained, and in the latter for microscopic examination of the particles dispersed in the liquid.

Filtration methods

Many factors are involved in the filtration of aerosols through porous media. The use of filters should aim essentially at enabling subsequent examination of the particulate matter to take place, but—apart from gravimetric measurements—such examination is often not easy, because of the difficulty of separating the matter filtered out from the filtration device.

One way of overcoming this drawback is to use soluble filtering media —for example, sugar or salicylic acid crystals—dissolving them in water and separating the particles retained by filtering the solution. However, it is essential to make sure that filtering media of this type (generally used in several layers) do not react with the particulate matter. Other filters used with good results include certain filter papers (ash-free), asbestos fibre or glass wool.

Recently, molecular filters, consisting of special plastic substances with very fine pores, have been introduced. These have the advantages of offering a minimum of resistance to the gas stream and also—when they are transparent—of being suitable for direct examination of the particle size, by determining the intensity of the light transmitted when the filters are moistened with oil having a suitable refractive index.

Such filters can also retain extremely small particles which would pass through practically any other filtering medium. Furthermore, they are soluble in acetone, making possible the practically complete recovery of the particulate matter in the laboratory and its subsequent analysis by other means, as required. Their sole disadvantage is that they lose their favourable properties when exposed to high-temperature gas streams.

Cyclone-type sampling

These devices are very suitable for sampling light ashes and dusts and can attain an efficiency of nearly 100 per cent. They have the following advantages: (*a*) simplicity of construction; (*b*) they can be made from materials resisting high temperatures; (*c*) they can be made to any size; and (*d*) the samples collected can be rapidly recovered.

The principle employed is to bring about a cyclonic or vortex movement of the gas or air to be sampled in a cylindrical container, so that the suspended matter is separated by centrifugal force.

Thermal precipitation methods

This method of sampling is based on the principle that hot surfaces repel particles suspended in the air, and that the particles thus repelled collect on cold surfaces. An airstream is made to cross the space round a heated filament situated between plates, with the result that the particles repelled by the hot filament settle on the plate. If the collecting plates are covered with colloidal matter, they can be directly examined under the electron microscope. The addition of an agitating device to the collecting plates helps to ensure a more uniform distribution of the particles which have been in suspension. The efficiency of such samplers is so high that it is not possible with any other sampling device to trap any particles which may have escaped.

Electrostatic precipitation methods

The principle employed here consists in submitting the gas stream to be sampled to the action of suitable electric fields so as to encourage the deposition of the particles in suspension on electrodes between which a high potential difference is maintained. In this case too, the efficiency may reach almost 100 per cent, but the samples obtained serve only for gravimetric analysis, since the particles are collected in a state of agglomeration which may be very pronounced.

Such devices are not suitable for sampling in the presence of explosive gases.

III. ANALYSIS

General

The atmosphere contains such an enormous number of substances, in concentrations which vary so greatly over the course of time, that continuous determination of its exact composition may be regarded as a problem which will remain very difficult to solve. The analyses which are made more or less daily concern only certain impurities known or suspected to be present in the air, and these represent only a small proportion of the total number of foreign substances dispersed in the atmosphere.

At the present time, there are not always analytical techniques available for dealing with the special forms and low concentrations of many atmospheric pollutants, but nevertheless even today it is possible to gain much useful information concerning a substantial number of pollutants by using analytical methods developed for other purposes.

It is the task of the analyst to obtain as much data as possible on the nature of the pollutants at the point selected for sampling. In some cases it may suffice to know the concentration of certain radicals or functional groups present in the sample. In other cases, however, apart from a knowledge of the chemical composition of the pollutants found, it may also be useful to know their physical nature, and consequently the sample should be collected in the form most suitable for the subsequent analytical investigation.

Since the aim of analysis in the special field of air pollution is to obtain information which can give an over-all picture, even if only an incomplete one, of the whole problem it is evident that certain requirements should be satisfied as regards concordance between the sampling and analytical techniques. These requirements are as follows:

(1) the efficiency of the sampling device used should be known as accurately as possible;

(2) the sampling device chosen should cause the minimum of change in the composition of the various substances to be detected;

(3) the apparatus should be sufficiently clean and the reagents sufficiently pure to give blank determinations which are lower than the lowest suspected concentrations of the substances sampled;

(4) the sampling method should supply the analyst with a quantity of pollutants within the quantitative limits called for by the sensitivity of the analytical technique to be used;

(5) the methods employed for the determinations should be as sensitive as possible and their repeatability should be satisfactory.

Other special difficulties are connected with the choice of analytical method, when there is not sufficient information on the conditions under which the compound to be examined may occur, since every analytical technique has its limitations, of which the most important are those relating to the concentration of the compound to be detected and the presence of interfering substances.

For all these reasons, a preliminary reconnaissance is advisable, using various analytical methods, such as microscopic methods, analytical methods based on the absorption of infra-red and ultraviolet light, emission spectrographic methods, X-ray diffraction and polarography—all of which unfortunately require, in addition to a specialized and skilled technical staff, special, high-precision equipment. On the other hand, the importance of the problem of air pollution is such as to warrant special consideration and the confrontation of all difficulties, no matter of what type they may be.

The very large variety of methods which have been applied, or which have possible application, to the analysis of atmospheric samples can be divided into three basic groups—namely, chemical methods, instrumental methods, and biological methods.

Since the second category is the one with which we are most concerned here, this report will deal only in general with the first and third groups, devoting more space to the field of analytical instrumentation as applied to atmospheric investigation. Of course, it will not be possible to describe in detail any particular instrument, for in view of the large range of instruments of all types on the market this would call for a separate examination of each example followed by a comparative study, which would require a special treatment beyond the limits of the present paper.

Analytical Methods

Chemical methods

In general, quantitative methods of chemical analysis can be subdivided into: gravimetric methods; volumetric methods (acidimetric and alkalimetric methods, oxidation and reduction methods, precipitation methods); colorimetric methods; nephelometric and turbidimetric methods; and chromatographic methods.

Gravimetric methods

Gravimetric methods of analysis are based on the principle of isolating the substance to be assayed in the form of one of its compounds which shows the maximum insolubility under the test conditions and has a known chemical composition. The weight of the compound is then determined and from this the quantity of the substance concerned can be calculated. Sometimes, however, the substances to be determined are already suitable for direct weighing so that in this case the assay involves only the direct determination of their weight. Gravimetric methods comprise the most simple analytical techniques, and can be used for:

(1) The determination of a gas (sensitivity approximately 1 mg). The methods may be applied to substances adsorbed on silica gel, activated charcoal or other adsorbents, by weighing the quantity of precipitate obtained with given reagents (as in the case of sulfur dioxide, which can be determined by weighing the sulfate precipitated as barium sulfate). Very large samples are usually required.

(2) The determination of dusts and soot collected in sedimentation jars following long sampling periods (of the order of 30 days for example) (sensitivity approximately 1 mg).

(3) The determination of particulate matter dispersed in the air (sensitivity 0.1 mg), sampled by collection on filter paper, by means of the impinger or electrostatic precipitators. Here too, rather large samples are necessary and the use of the micro-analytical balance may be required.

Volumetric methods

In volumetric analysis, the components are not weighed but determined by means of reagents of known concentrations. As has been mentioned, these methods include:

(1) Alkalimetric and acidimetric methods for the determination of bases and acids, using solutions of an acid or a base, respectively, in the presence of indicators which undergo a colour change according to the alkalinity or acidity of the medium. (Substances used as indicators are electrolytes, generally weak acids or bases and in the undissociated state—that is, when they are electrically neutral—their colour should differ from that of their ions.)

(2) Oxidation and reduction methods which are based on the oxidation or reduction of the compound to be analysed by means of a standardized substance, the quantity of oxygen given up or consumed being measured according to the colour change developed in the reaction, in the presence or absence of an indicator.

(3) Precipitation methods (suitable in particular for assaying the halogens) in which use is made of silver nitrate solutions of known strength which precipitate the halogen in the form of silver halides.

Acidimetric and alkalimetric methods can be used for the volumetric determination of acid and basic substances and are very sensitive, being able to detect less than 0.01 p.p.m. by volume of the gas stream sampled. They call for large samples. Examples of these methods are the determination of sulfurous anhydride, oxidized to sulfuric acid, and of ammonia, by means of direct titration with standard solutions of sodium hydroxide and sulfuric acid respectively.

Other gases can be determined with a sensitivity exceeding 0.01 p.p.m. by volume, by means of iodometry (oxidation and reduction), particularly in atmospheres of known composition, using samples obtained by absorption, as in the case of sulfuric anhydride, chlorine, aldehydes and oxidizing agents. For the last mentioned, this is perhaps the most sensitive and advisable method.

Finally, the halogen acids in particular (hydrochloric acid, hydrofluoric acid, hydriodic acid) and their salts can be assayed by precipitation, but the reaction is not sufficiently sensitive to be of wide application.

Colorimetric methods

Colorimetric methods are on the whole the most satisfactory type of determination among the various analytical procedures known today. A very large number of elements, radicals and organic compounds form coloured complexes with special reagents, and such colorimetric reactions are sometimes sufficiently sensitive to detect even the most minute traces of the compounds which are being sought. Naturally they call for the use of suitable and more or less sensitive apparatus for determining the intensity of the coloration produced, such as visual, filter or other types of photometer, photo-electric apparatus using photo-cells or photo-tubes, indicators or recorders. This might therefore seem to be more a branch of instrumental analysis, but since the colorimetric techniques have entered into very extensive use they can now be considered as simple chemical methods. This is all the more true in that they involve only the instrumental measurement of actual and specific chemical reactions, and sometimes also normal titrations with special indicators. They can be used for the analysis of gas and particulate matter, sometimes with a sensitivity of less than 1 μg, but occasionally they require large samples and difficult separation and concentration techniques for the compounds to be examined.

The colorimetric tests include some of the best assay methods, particularly in the analysis of metallic vapours or dusts, formaldehyde, oxides of nitrogen, ammonia, chlorine, hydrogen sulfide, fluorine, carbon monoxide, carbon dioxide, as well as many other organic and inorganic compounds. Sulfurous anhydride, for example, can be assayed with a sensitivity of 0.001 mg, by measuring the violet colour produced by the reaction of benzidine sulfate with N-1-naphthyl-ethylenediamine hydrochloride.

Nephelometric and turbidimetric methods

Nephelometric and turbidimetric methods involve procedures in which the precipitates, obtained by means of specific reactions, are quantitatively assayed by optical measurement of light passing through the corresponding suspensions. They are suitable for the determination of gas and particulate matter, do not require specially large air samples and find their best application in the determination of sulfurous anhydride (by means of precipitation as barium sulfate and estimation of the turbidity produced by the precipitate with a sensitivity of 0.005 p.p.m. by volume) and of chlorine (which is precipitated as silver chloride, likewise with a sensitivity of 0.005 p.p.m. by volume).

Chromatographic methods

Chromatographic methods make possible the separation and identification of many substances by means of adsorption on adsorption columns, or on specially sensitized paper, followed by the elution of the various compounds adsorbed with selective solvents. In this way, the components of complex mixtures can be separated without undergoing any chemical change. Traces of inorganic ions can be detected by such methods with extreme accuracy and with considerable saving in time and material as compared with other methods.

The sensitivity of such methods is very high (sometimes less than 1 μg) and furthermore they are suitable for any kind of substance. They are particularly useful, however, for metals and volatile organic compounds. Their application to air analysis is still in the initial stages although they are already being extensively used in special cases—for example, the determination of benzpyrene in urban atmospheres.

Instrumental methods

In order to ensure analytical specificity, selectivity and sensitivity, methods are required which are based on clearly defined physical principles, as well as apparatus designed for the exact application of such principles.

In general, instrumental analysis is the application to analytical chemistry of special physical and physico-chemical methods. Such methods are finding an ever increasing application because of the extreme rapidity with which they enable certain types of analysis to be carried out (otherwise too long and complex if the ordinary methods are used) and because of their efficiency in cases where the classical methods generally give unsatisfactory results. In principle, however, it is not possible to make a fundamental distinction in classification between instrumental and non-instrumental methods. Even the simplest of the chemical methods—namely, gravimetric determination—also calls for the use of an instrument—the balance—which, although its employment is of a routine and elementary nature, nevertheless remains an instrument. Furthermore, as we have seen, colorimetric, nephelometric and turbidimetric determinations also require the use of sensitive instruments for assessment of the intensity of the colour or turbidity (produced by given reactions), by measurement of the light absorption caused by the particles suspended in a liquid (turbidimetry) or measurement of the intensity of the light dispersed by a suspension of particles in a liquid medium (nephelometry).

Determinations of pH and salinity are sometimes necessary in the analysis of air, using samples obtained by absorption of the gas phase under study in the simplest of absorbing media—namely, water—so as to determine, for example, the concentration of certain acid or alkali mists or the amount of salts, such as sodium chloride, in suspension. Potentiometric titration is used, for instance, in the analysis of atmosphere containing sulfides or mercaptans, and in this method the indicator is replaced by the electromotive force developed between a reference electrode and a measuring electrode.

Both these methods form part of a group constituting that branch of analytical chemistry which is known today as "electro-analytical chemistry".

This latter makes use of the phenomena which occur in an electrochemical cell (that is, a cell consisting of two metal electrodes—of the same or different metals—immersed in the same or two different electrolytes and connected to apparatus suitable for the exchange of electric energy with the cell). According to the particular case, the behaviour of the cell is determined by measuring the various electrical quantities involved, namely:

(1) the amount of electricity flowing in the circuit of the electrochemical cell (coulombometric method);

(2) potential difference in the electrochemical cell (potentiometric and potentiometric titration methods);

(3) current flowing in the electrochemical cell circuit (amperometric and amperometric titration methods);

(4) current-potential difference (voltametric methods, which include the polarometric methods);

(5) resistance, or better its converse, conductance, of the electrochemical cell (conductometric and conductometric titration methods);

(6) dielectric constant (high frequency methods);

(7) other methods (electrogravimetric, electrolytic separation methods, etc.).

In view of the fact that optical determination methods, in colorimetric, nephelometric and turbidimetric analysis, as well as the electrochemical methods, are in constant and routine use as auxiliaries to the normal methods of chemical analysis, we shall describe in this chapter only those instrumental methods (and the fundamental information they provide) whose complexity is such that they still represent special means of research, not yet in routine use, but whose application to the field of air pollution can bring about significant advances in our knowledge and in the solution of the problem itself. These methods are the following: emission spectrometric methods; absorption spectrometric methods (spectrophotometry) in the ultraviolet and infra-red regions; X-ray diffraction methods; mass spectrometric methods; polarometric methods (polarography); methods employing microscopy; refractometric methods; combustion or thermal conductivity methods; other instrumental methods (radioactivity, sound absorption).

We shall deal in a separate section with automatic (recording) apparatus for sampling and analysis.

Emission spectrometric methods

The general definition of spectrometric methods includes all techniques based on the phenomena of emission or absorption (and possibly the related fluorescent re-emission) of electromagnetic radiation in the visible, infra-red and ultraviolet regions of the spectrum. For obvious reasons, we shall not deal with spectroscopy—although this is first in chronological order—and shall limit ourselves to spectrometry and in particular to spectrometric emission methods. The fundamental principle upon which such analytical methods are based is that any element, when in an excited state, has its own special emission spectrum, so that emission spectrometry is widely used to determine the composition of mixtures or compounds and is indispensable in the exploratory examination of particulate matter.

Emission spectrometry can be applied to the analysis of more than 70 of the known elements. The alkaline and alkaline-earth elements can be detected with the highest attainable sensitivity—namely, down to 0.1 p.p.m. by weight of the quantity of substance examined—while some 40 - 50 other elements can be detected with a lower degree of sensitivity, ranging from 10 to 1 p.p.m. by weight. This analytical technique, by now well established, requires high precision apparatus for its application. Such apparatus consists of: (*a*) a source of radiation; (*b*) a means of splitting up the radiation, which may be either a prism or a grating; and (*c*) a receptor, which can be either a photographic plate, as in the case of the older spectrographic equipment, or a series of photo-tubes, as in the more modern spectrometric apparatus.

Spectrometric analysis (quantitative) involves the spectroscopic characterization (qualitative) of the elements, based on identification of the characteristic lines in their emission spectrum. Such identification can be made by means of a comparator microscope for measurement of the distance between the lines themselves, while quantitative determination is based on the consideration that the energy emitted for a given spectral line of an element is proportional to the number of excited atoms and consequently to the concentration of the element in the sample.

In air analysis, spectrometric emission methods are applied mainly in research work, especially for the identification of the elements present in particulate matter. The method is particularly indicated for beryllium, sodium, potassium, calcium, lead, cadmium and antimony.

Absorption spectrometric methods (spectrophotometry)

Spectrophotometry, based on the absorption of a certain range of electromagnetic radiation in the visible, ultraviolet and infra-red regions of the spectrum, is one of the most important of all instrumental methods of analysis and is consequently of potential value for the analysis of impurities in the atmosphere. Absorption in the visible spectrum is the basic principle of the methods of measurement applied in colorimetric and turbidimetric analysis, which, as has been seen, can be applied to the analysis of both gaseous and particulate matter. These methods are frequently sufficiently specific, although it is sometimes necessary to isolate and concentrate the compound to be examined so as to avoid interference by other compounds.

The absorption varies with the wave-length of the incident radiation, so that measurements should be carried out either with monochromatic light or with light having a limited spectrum, which can be produced by using special light filters. The wave-length of the beam emerging from the source of radiation is suitably adjusted in this way and then passes through a layer of the substances to be examined, while the intensity of the non-absorbed radiation is determined with a suitable detecting apparatus.

In the visible region of the spectrum, the concentration of a coloured substance analysed spectrophotometrically can be derived from the Lambert-Beer relationship, according to which light absorption is proportional to concentration, when the layers analysed are of equal thickness. However, spectrophotometric methods can also be applied to the analysis of colourless substances by studying absorption in the ultraviolet region of the spectrum, but this method is not satisfactory for the analysis of compounds present only in traces.

The difficulties in practical application to atmospheric analysis are thus inadequate sensitivity and the interference arising from the presence of numerous other substances in the atmosphere. Nevertheless, good results can be obtained in the analysis of impurities in industrial atmospheres and in gaseous effluents when the determination is carried out directly on the gas stream or when suitable sampling devices are employed after the gas has been washed. In general, such methods are suitable for organic substances, in particular benzene and aromatic compounds. Infra-red spectrometry is more successful, on the other hand, since it can be applied to either solid, liquid or gaseous substances.

So far, most of these applications have been restricted to the field of research, but it is to be hoped that these techniques will have wider practical application, in view of the following advantages:

(1) all the molecules of a substance in the solid, liquid or gaseous state, as well as in solution, have a characteristic infra-red spectrum;

(2) a single spectrogram can serve to identify at one and the same time: the radicals, the elements with which they are substituted and their position in the molecule;

(3) the spectrum shows both the nature and the quantity of the substance being analysed;

(4) this gives an indication of the degree of purity of the sample prepared;

(5) the techniques used are very simple;

(6) only relatively small samples are required.

However, it should also be borne in mind that:

(1) the sharpness of the spectrum decreases with increasing molecular weight;

(2) in the present state of this technique, we have not sufficient knowledge to classify all the possible absorption bands, so that in many cases pure, standardized reference samples are necessary;

(3) traces of impurity show up only if their absorption is high;

(4) dilute solutions are difficult to analyse unless the substances present are strongly absorbent.

Samples for infra-red spectrophotometric analysis often call for considerable preliminary chemical manipulation and, with few exceptions, natural, untreated gas samples are not very suitable. The impurities must often be concentrated or isolated by means of absorption or cooling, or a combination of both, and in this way exceptionally high sensitivity can be attained, as in the case of ethylene, which can be detected in concentrations as low as three parts per thousand million of air by volume.

Other substances present in the atmosphere to which this type of analysis can be applied are: (*a*) mixtures of esters, aldehydes or alkalis, sampled by adsorption on charcoal, followed by condensation using an ice-salt cooling mixture or dry ice, and subsequent extraction with carbon tetrachloride; (*b*) chlorinated hydrocarbons, sampled by fractional condensation—benzene and hydrocarbons; and (*c*) carbon dioxide, carbon monoxide, hydrocyanic acid, water vapour, ozone.

Still in the field of spectrophotometric analysis, it is possible to make use of a phenomenon shown by certain substances—namely, the re-emission of the light energy absorbed in the form of a fluorescence, whose intensity is measured. By means of this measurement of fluorescence it is possible to analyse trace amounts of certain kinds of atmospheric components, but it is sometimes necessary to eliminate interfering substances so as to ensure the specificity of the determination.

The sensitivity obtained with this method is often less than 1 μg. To date, it has been used for the detection of beryllium, antimony and hydrofluoric acid.

X-ray diffraction methods

These are suitable for the qualitative and quantitative analyses of crystalline substances, such as certain mineral dusts which can be present as a suspension in the atmosphere. However, X-ray diffraction methods should be considered rather as analytical procedures ancillary to other forms of analysis, such as emission spectrography or the methods of analytical chemistry. It is of particular interest that the samples used for such analyses can be recovered and used for other determinations employing other techniques.

X-ray diffraction methods also form part of the spectrometric methods, but while the latter involve measurements ranging from a wave-length of 40 μ (extreme limit of the infra-red) to 0.2 μ (extreme limit of the ultraviolet) X-ray diffraction measurements involve wave-lengths of the order of 0.0001 μ. The sensitivity of such methods thus arises from the great penetrating power of X-rays, also as regards solid bodies which are opaque to any other form of radiation.

It is known in fact from mineralogy that crystals can serve as diffraction gratings in the study of X-rays and that each crystal has its own particular diffraction pattern, so that it is possible to identify unknown substances by comparing their diffraction patterns with those of known substances. Quantitative determinations are sometimes possible, based on measurement of the intensity of the diffraction lines after having photographed them on a film. Nevertheless, these methods are mainly of qualitative interest since their quantitative sensitivity does not exceed one per cent of the substance examined. Moreover, they require very large samples and considerable practical experience on the part of the investigator. They are employed principally for the detection of quartz crystals in the air.

Mass spectrometric methods

Mass spectrometry is based on the principle that if a gas is ionized by means of electric discharges, and if the collimated beam of positive ions produced is subjected to the simultaneous action of an electric field and a magnetic field at right-angles to each other, then the ions are deflected from their initial straight-line direction of propagation and become distributed along a curve, the deflection being proportional to the mass of the ions. The result is a mass spectrum, and the apparatus which records it on a photographic plate bears the name of " mass spectrograph ", while a " mass spectrometer " is an apparatus in which the radii of curvature of the paths of the various ions can be altered by changing the speed of the ions or the intensity of the magnetic field. In this way the nature and the quantity of the various kinds of ion can be determined by measuring the current flowing in a collecting circuit when the radius of the paths is changed.

Mass spectrometry has recently aroused some interest in connexion with the atmospheric pollution caused by petroleum refining, because of its rapidity and satisfactory sensitivity at the concentrations of waste gas involved. Naturally, this method calls for a wide knowledge of mass spectra in order to make comparisons and identifications. The samples need not be very large, but it is essential to isolate and concentrate the fractions to be examined and to convert these into gaseous form.

The method can be used for the identification of toxic gases with a sensitivity of 100 p.p.m. by volume with samples of only 50 ml of air. The sensitivity can be considerably increased by using samples concentrated by cooling, and in this way a sensitivity of 0.001 p.p.m. by volume can be reached in the case of certain compounds. The method is useful in particular for hydrocarbons and compounds of hydrocarbons with chlorine, nitrogen and oxygen.

Polarographic methods

Among the electrochemical methods of analysis listed at the beginning of this chapter must also be mentioned one making use of the chemical phenomena which occur in an electrochemical cell, and based on interpretation of the ratio between current and potential difference. This method is generally referred to as voltametry or polarography, in the particular case where a dropping mercury electrode is used. In the polarograph, the voltage applied to the cell is regulated by means of a potentiometer and the current which passes through the cell is read on a galvanometer. These methods are applicable to the determination of any substance which can be reduced or oxidized, so that they can be used to identify a large number of organic and inorganic compounds.

To obtain the maximum sensitivity and specificity, various chemical treatments are necessary in order to isolate and concentrate the sample, although the simultaneous determination of several compounds is sometimes possible. Substances such as the following can be analysed polarographically: antimony, arsenic, benzene, bismuth, cadmium, chromium, cobalt, copper, formaldehyde, lead, manganese, nickel, oxides of nitrogen, thallium, tin, titanium, vanadium, and zinc.

Furthermore, it is possible to distinguish between a certain number of aldehydes and ketones (formaldehyde, acetaldehyde, butylaldehyde, crotonaldehyde, acetone, methylisobutylketone, cyclohexanone) in semi-carbazide solution. A sensitivity of 0.1 μg of aldehyde per ml of solution can be attained.

Methods using microscopy

The use of microscopy in air pollution research is limited to the examination of particulate substances of all types for counting the particles themselves, measurement of their size, and optical observation of their surfaces. In this way the following can be examined: tarry matter, small drops of oil or water, crystals such as quartz and ammonium sulfate, nitrates, sulfates, halides, and ammonium carbonates.

Microscopy is sometimes combined with chemical procedures for the preparation of precipitation samples and their crystallization. According to the type of apparatus used, particles with diameters as small as 0.2 μ can be detected, and with the use of the electron microscope, which extends sensitivity to as low as 50 Å = 0.005 μ, it is possible to study even smaller particles present in the atmosphere. While the non-electron microscope gives quantitative results only as regards the number and size of the particles, the electron microscope, although requiring a difficult and special preparatory technique, can yield an absolute analysis of particulate matter, since such apparatus can also be used for X-ray diffraction measurements with very small amounts of substance.

Refractometric methods

These are based on the use of the refractometer for determining the refractive index of the substance under study. The technique is of limited application in air analysis, because of its lack of specificity and sensitivity. It can be used solely under conditions in which only one or two known vapours or gases are to be determined whose refractive indices are sufficiently different from that of air to permit a certain degree of accuracy at the lower concentrations.

Refractometric methods can be used for the analysis of emanations from waste gas pipes and in areas very close to the source of pollution. In this way, variations in the refractivity of the air due to an unknown proportion of vapour or gas present in it can be determined and, on comparing these values with the unit of change in refractivity—that is, the change caused by the presence of 1% of vapour or gas in the air at 25° C and 760 mm Hg (using standardized samples for the various substances)—the percentage present of the substance concerned can be determined.

Thermal conductivity methods : combustion

Since the measurement of the thermal conductivity of gases and vapours cannot serve to identify the gas, but is only a method for evaluating the concentration of known substances, procedures based on such measurements do not have a very wide application. They give only an over-all index of the thermal conductivity of a gas mixture and consequently can only be applied to those cases where the conductivity can be measured before and after eliminating a particular constituent, so that the latter can be determined from the difference.

In certain cases, however, it is possible to use a combustion apparatus known as an explosimeter, which gives the concentration of vapour or gas as a percentage with respect to the minimum explosion limit. In this method, a filament is electrically heated in special apparatus to a temperature at which the gas or vapour becomes oxidized and burns at the surface of the filament. In other cases, chemicals are used to bring about catalytic oxidation of the gas. The energy developed during oxidation (combustion) is then measured with a thermocouple. This is the method most used for determining carbon monoxide, but even in this case there is the difficulty of dilution.

Such methods are consequently of little value for work on air pollution and, as had been said, are limited to the industrial analysis of gaseous effluents.

Other instrumental methods

Radioactivity. Analytical methods based on the measurement of radioactivity have also been used in atmospheric analysis to detect substances and impurities present as traces. There are two basic methods for the measurement of radioactivity, deriving from the dilution and activation techniques, respectively.

In the first case—that of dilution—the element to be detected is diluted with one of its radioactive isotopes and the specific activity of the diluted substance is subsequently measured with a detector. This method calls for the isolation of a measurable amount of substance and is considerably upset by impurities and by samples which are not well selected. As regards air pollution, it is used practically only to measure the efficiency of smoke filters.

The second method—activation—is perhaps the most sensitive of all known analytical methods. It consists in the artificial transformation of the substances under examination into their radioisotopes and subsequent measurement of the quantity of disintegration product. This method is not subject to interference, because of the invariability and specificity of the half-life periods of the radioisotopes. However, as always in complex analytical methods using instruments, the greatest difficulties are those involved in the sampling, manipulation and preparation of the samples, apart from the problem of obtaining technical personnel with sufficient experience in this field of analysis and research.

Sound absorption. This method of analysis is used for measuring particle size. It is based on the fact that particles subjected to the influence of a sound field of a particular frequency are affected by those vibrations whose amplitude is related to their size. The instrument used for this purpose consists of a sound generator which can be adjusted to the desired frequency, a thin observation cell, towards which the sound is directed and into which the sample is introduced, combined with a lighting system and a means of observation or photographic recording. The photograms obtained show the paths corresponding to the vibratory movements of the particles, and by measuring them the particle sizes can be determined. However, very dilute samples are necessary, since only a few particles should be present in the cell if clear and interpretable photograms are to be obtained.

Conclusions

From the preceding it can be seen how the same substance can be determined by a variety of methods. It is the task of the analyst to make a critical comparison of the various possible methods which can be applied. Some procedures are very suitable for routine work and not practicable for research work, and vice versa. No precise rules can be laid down for the choice of the method and means of analysis, except that those procedures should be selected which give the best " accuracy ", and highest " sensitivity ", the highest " selectivity " and the best " precision ", without forgetting that rapidity of analysis is also an important factor.

The " accuracy " is the sum total of the errors—absolute or relative—which can influence the results of an analysis. It should not be confused with " precision ", which represents the repeatability of the experimental results. " Sensitivity " expresses, as an absolute quantity, the minimum amount of substance which can be analysed, or the minimum analysable concentration in a solution or in any other medium of dispersion. The " selectivity " of a method is shown by enumerating those substances which interfere with it.[1]

It is difficult to find a combination of all these properties in their highest degree in one and the same method. However, it is probable that by combining a wide range of techniques optimum results can be obtained. In the field of air pollution, nevertheless, there is always the inherent difficulty of sampling, as already discussed.

Continuous recording instrumental methods

Under certain circumstances, calling for repeated analyses in order to determine the behaviour of a source of pollution and study its effects with respect to time, automatic, continuous recording apparatus are of value. These instruments combine the functions of sampling and analysis in the same apparatus. Although such apparatus is not usually extremely accurate, nevertheless it furnishes graphs which can be of great value in directing the research worker towards more-specific tests employing discontinuous sampling and analysis, under the conditions indicated as the most interesting ones.

Apparatus of this type is constructed on the basis of the chemical, electrochemical, optical and other principles already enumerated. It can be applied for research both on gaseous substances and on particulate matter.

In this field, most attention has certainly been directed to sulfurous anhydride among gaseous impurities, since it is the most widespread and the one which perhaps has given rise to most incidents and disputes. One of the most widely used automatic analysers for the determination of sulfurous anhydride is the Thomas autometer, which measures the electrical conductivity of the sulfuric acid solution produced by the oxidation of sulfurous anhydride following absorption in slightly acidified water containing hydrogen peroxide.

There are two versions of this apparatus, an accumulating and an instantaneous model. The accumulating types record conductivity continuously for absorption periods of 30 minutes, while the instantaneous types record the concentrations at two-minute intervals. Both types can be combined into a third or " mixed " type, in which the concentrations are determined at intervals of two minutes, as well as the average concentrations over 30-minute intervals. Since, however, many gases or aerosols yield an electrolyte in solution, such apparatus measures the total pollution and not only that specifically due to sulfurous anhydride, although the total is expressed in terms of the latter.

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[1] It should be explained in this connexion that both in the foregoing and in the following pages, as can easily be seen, the figures for sensitivity are expressed sometimes by weight and sometimes by volume. In the first case this means, as previously mentioned, the minimum quantity of substance, as an absolute value, which can be analysed, while in the second case it means the minimum concentration of substance which can be detected in a gaseous mixture, whether waste gas or the atmosphere. Thus the expression " parts per million " indicates the number of cubic centimetres of substance under examination per cubic metre of gaseous mixture, under normal conditions of temperature and pressure—i. e., 0° C and 760 mm Hg.

Another apparatus, the titrilog, also measures sulfurous anhydride down to a concentration of 0.02 p.p.m. by volume, by means of oxidation of the gas stream in an electrolytic cell in the presence of bromine. Again, this apparatus is sensitive not to sulfurous anhydride alone but also to many other gases which can be oxidized, such as hydrogen sulfide, mercaptans and unsaturated hydrocarbons. On replacing the bromine by a starch-iodide solution and titrating the oxidizable substances photometrically, better selectivity is possible, since iodine is less reactive than bromine with such substances. Even in this case, however, it is not possible to eliminate all interference, at least as regards hydrogen sulfide.

With suitable modifications, however, such apparatus can be used in a more selective manner, so that the titrilog can also give values for hydrogen sulfide and the mercaptans and the autometer can detect any substance which gives an electrolyte in water, either directly or after combustion. Thus, the autometer can be used to determine hydrogen sulfide with a sensitivity of 0.01 p.p.m. by volume, following the production of sulfurous anhydride by combustion of the hydrogen sulfide on a platinum filament electrically heated to 550° C. The same apparatus, with lower selectivity and sensitivity, however, can also be used for measuring the amount of hydrofluoric acid present in the air.

For the latter compound another analysing apparatus has been developed with a sensitivity of 0.1 parts per thousand million by volume of hydrofluoric acid, which records the quantity of hydrofluoric acid by measuring the fluorescence which the latter produces with certain magnesium salts. Other equipment can give the concentration of hydrogen sulfide by measuring the variations in the intensity of light transmitted through plugs impregnated with lead acetate during the passage of a gas stream containing hydrogen sulfide. It is possible in this way to cover concentrations ranging from 0.1 to 500 p.p.m. by volume.

Recording analysers are also applied for the analysis of oxidants (ozone, oxides of nitrogen, chlorine). Such apparatus measures electrometrically or photometrically the amount of iodine liberated from solutions of potassium iodide by the oxidants.

An automatic instrument, the microsensor, can detect and measure many kinds of gaseous impurity in amounts as low as 0.1 μg/l. It consists of a colorimetric comparator which measures the colour changes produced on paper sensitized with special reagents. It is intermittent in action, with cycles varying according to the speed of reaction of the substances to be detected with the sensitizing reagents. In general, these cycles do not exceed 10 seconds in length.

Particulate matter can also be dealt with by recording apparatus. In these instruments, the intensity of light dispersed in the atmosphere owing to the presence of particles is measured and the visibility recorded directly. It is possible to relate these measurements of opacity to the particle concentration and size, following suitable calibration of the apparatus. The same principle—namely, measurement of the intensity of the light dispersed in the atmosphere owing to the presence of particles—is also made use of in apparatus counting the number of particles present, although the sensitivity of such measurements is rather low. The electrostatic principle is also applied to the continuous counting of particles, by measuring the impulses produced by the particles settling on the metallic filament which acts as precipitator. Instruments of this type are sensitive to particles 2.5 μ or more in diameter.

A modification of the autometer already mentioned makes it possible to record the quantity of sulfuric acid present in the air in the form of aerosol. This apparatus is provided with an electrostatic precipitator consisting of a rotating stainless steel disc which collects the sulfuric acid droplets and is washed by a continuous stream of water. In this way, a solution is formed whose conductivity is measured electrolytically. Naturally, this apparatus also measures any other acid mist and can be specific only if placed near a known source.

Another example of a sampler and analyser is a monitor for atmospheric radioactivity, in which the aerosol is deposited electrostatically on the sampling device, connected to a continuous recording radiation counter.

Instrumental methods for the control of radioactivity

Among the causes of air pollution, those arising from radioactivity are becoming increasingly important, both because of the growing number of industries employing radioisotopes and because of the development of nuclear plants (both for power and for research), with the consequent production of fissile materials, decontamination of waste matter, etc.

Ionizing radiation causing pollution can vary very widely in type—that is, electromagnetic and corpuscular radiation, comprising alpha rays, beta rays, gamma rays, X-rays, proton beams, electron beams, slow neutrons and fast neutrons. Exposure to such radiations may cause considerable harm to living tissue, so that it is becoming increasingly necessary to employ suitable detectors in order to afford some protection against the possibility of harm of this kind. The problem is so important, and the reliability of the indications given by these detectors is such that occasionally, particularly in the case of measuring instruments installed near sources of pollution, they are fitted with alarm signals which are set off whenever the radiation level exceeds—in the zone under control—the danger limit which has been established for the various forms of radiation. The problem is rendered particularly complex by the uncertainty of the definitions of the measuring units (both fundamental and derived) used for radiation and of the basic concepts related thereto. The physical nature and intensity of the radiation also present other difficulties, as does our still very slight knowledge of the results of the interaction between radiation and matter and its effects on the living cell and tissues.

Among measuring instruments in this field (including pocket control and safety detectors for plant personnel) the following may be listed: ionizing chambers; Geiger-Müller counters and proportional counters; scintillation detectors; photographic detectors; chemico-colorimetric indicators; and colorimetric measuring instruments.

Biological methods

Effects on plants and animals

It is often possible to identify a pollutant and a source of pollution by analysis of vegetation and of the body fluids of animals bred in the infected area. In certain cases, the tissues and organs of dead animals are very useful for the identification of toxic substances and the interpretation of the analytical results obtained. When, in fact, the concentration of a pollutant remains below the toxic level, there is no way in which its effect on plants and animals can be directly observed. However, when these limits are exceeded, harm is visibly caused to plant and animal life, according to the chemical nature of the pollutant, the time of exposure and climatic conditions. Sometimes the effects caused can be so specific as to serve as a sufficiently sensitive means of determining the nature of the contaminant. Complications may be introduced, however, owing to the concomitant effects of a variety of substances.

Biological studies of the effects on plant and animal life suitably exposed to the polluted atmosphere can be useful as regards both gases and particulate matter. Many tests are extremely sensitive and specific: beryllium can be detected in amounts as low as 0.0003 μg and lead in as little as 0.01 μg. Such tests may require very large samples, however, and often call in addition for the isolation and concentration of the components.

Sensory tests

Finally a certain number of substances can be detected because of their organoleptic properties; odour, lacrimation and irritant effects on the respiration can be important factors, making it possible to assess the extent of pollution. In such cases, nevertheless, quantitative evaluation is seriously limited by the many subjective and physiological influences to which perception is exposed. The sensitivity of such methods is consequently very limited and they should be reserved in particular for qualitative research or reconnaissance, and be entrusted to specially " sensitive " personnel, trained for the recognition of given substances on the basis of their organoleptic properties.

IV. CONCLUSIONS

As mentioned in the introduction, this report represents a preliminary contribution towards a selection from among the available material of whatever may seem most suitable for attaining a more limited range of tolerance. This is necessary in view of the infinite variety of special conditions and circumstances applying to the use of sampling and analytical apparatus and methods in the field of air pollution. In this paper, an attempt has therefore been made to select sampling methods and to give a critical review of analytical procedures in use at the present time, or potentially suitable for use, in the various laboratories of countries interested in the problem of air pollution.

However, as was foreseen and as has been mentioned, considerable difficulties were encountered from the outset in the classification, selection and interpretation of sampling and analytical methods and techniques. These difficulties are of some importance as regards the problem of ensuring that the presentation is universally understandable. From the outset they have arisen from a lack of uniformity among the various authors in describing their work—a lack of uniformity which is not limited moreover to nomenclature, but also extends to the presentation and interpretation of the data. For this reason it would seem advisable to conclude this report by giving a few suggestions, with the aim of eliminating, as far as possible, all uncertainty in the presentation, reading and interpretation of future contributions in this field. If these suggestions were followed it would be possible, without an effort out of proportion to the results, to ensure the highest degree of uniformity attainable in this field.[1]

(1) Definition of the terms and expressions used (and creation of any which may be necessary) seems essential in order to establish a glossary of terms relating to air pollution. This glossary, which could subsequently be converted into a multi-lingual international dictionary, would give a definite and authoritative interpretation of all words or expressions, and any circumlocutions which today may be necessary to indicate special circumstances and cases could be translated into precise and logical terms having an absolute, international significance. In this way it would be possible, in time, to create a technical and scientific language applying specifically to the huge field of air pollution and eliminating all words and expressions which —although idiomatically correct in the various languages—are not scientifically " precise ", this adjective being derived here from the noun " precision "

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[1] It should be pointed out that standardization in the field of air pollution is being promoted by the Commission on Toxicology and Industrial Hygiene of the International Union of Pure and Applied Chemistry, and that the American Society for Testing Materials Committee D-22 has developed a programme for standard air sampling methods.

as used in chemical analyses, when it expresses the repeatability of experimental results.

(2) After having studied the large volume of work carried out in the fields of sampling and analysis as related to air pollution, and after having briefly explained the principles and the nature of the information supplied by a selection of the various methods and techniques employed, it appears urgently necessary to attempt to standardize the latter, so as to avoid any doubt or uncertainty in the comparison of experimental results from different sources.

The American Public Health Association, the American Water Works Association and the Federation of Sewage and Industrial Wastes Association have compiled a selection of standardized methods for the examination of water, sewage, industrial wastes, etc. This outstanding publication,[1] which has now reached its tenth edition, is still so young that it continues to include (and can hardly avoid doing so in future) methods indicated as under test —that is, methods which have not yet been sufficiently tried out and tested to be put forward as standard.

The proposal which we wish to make, therefore, is that of drawing up as soon as possible an initial list of sampling and analytical methods which might be termed "for guidance". The various workers could, in the course of time, contribute the experience, controls and tests necessary to transform this list of methods "for guidance" into standard methods which, in addition to giving experimental details, would also state their efficiency, accuracy, precision and selectivity, with respect both to sampling and to analytical techniques.

It must be recognized, however, that such a task would involve considerable difficulties, because of the widely divergent conditions and circumstances under which the qualitative and quantitative examination of polluted atmosphere and of the open air may have to take place. These conditions and circumstances also exist in other fields, such as that of water supply, but on a much smaller scale, both numerically and quantitatively, so that it is easier to classify, prevent or overcome them. The initial work suggested as a basis should not be postponed because of this, however, since it could well be a source of improvements and developments in our knowledge of this particular field to an extent which might seem incredible at the present time.

(3) The presentation of analytical results should be made "uniform", for in this field one is really confronted with a kind of babel, quite apart from differences between units of measurement, whether decimal or otherwise. Side by side with "grams per litre" and similar expressions, we find "percentage by weight" and "percentage by volume", "parts per

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[1] *Standard methods for the examination of water, sewage and industrial wastes*, 10th ed., 1955, Baltimore, Waverly Press.

million by weight " and " parts per million by volume ", as well as other mixed expressions. In addition to results converted to " 0° C and 760 mm Hg ", there are others expressed at " 25° C and 760 mm Hg ", as well as still other intermediate forms. Finally, sensitivity of the various methods, as well as being expressed in absolute figures, is expressed in relative values, and so on.

Results are expressed in so many and such diverse ways that usually the conversion of any considerable number of them, from one form to another, for purposes of comparison, becomes an arduous task, sometimes economically impracticable for reasons of time. Again, it should not be forgotten that there are many papers in which the data are reported without any reference to the units of measurement employed. All this shows how necessary, if not indispensable, is the standardization of the way in which results are expressed. Tables for the conversion of one form of presentation into another are not sufficient; it is also necessary for the standardized methods suggested above to employ definite and constant units of measurement. This would lead to an enormous saving of time and greater clarity in the results, as well as to an accurate technical and scientific picture of the possibilities and limitations of present and future research in the field of air pollution.

Morris KATZ, M.Sc., Ph.D.*

SOME ASPECTS OF THE PHYSICAL AND CHEMICAL NATURE OF AIR POLLUTION

Introduction

Within recent years attention has been drawn to the complexity of the problem of air pollution in large urban centres. Although information is being rapidly acquired on many constituents of the contaminated atmosphere, the number of these contaminants is still unknown. The concentrations of most atmospheric contaminants are extremely low and the accurate determination of such substances taxes the ingenuity of the analytical chemist. All the activities of man, the biological processes of living matter and natural phenomena upon or below the surface of the earth result in the liberation of gases, vapours, dust and aerosols of suspended particulates to the atmosphere. The study of the nature, properties and behaviour of these substances represents a challenge to the competence and skill of physicists, chemists and biologists.

It is almost an axiom that some fraction of each component of the enormous number of raw materials and products of industrial and public activities finds its way into the atmosphere as a contaminant. Gases and vapours thus liberated may pollute the atmosphere to the extent of a few parts per million or of as little as one part per thousand million or less. Nevertheless, the toxicity to living matter of some compounds in the parts per thousand million range may be greater than that of others which pollute the air in concentrations that are a thousand-fold greater. Aerosol contaminants usually range from less than 100 $\mu g/m^3$ in relatively clean air to over 500 $\mu g/m^3$ in urban areas of comparatively heavy pollution. During smog periods the concentrations may rise above 1000 $\mu g/m^3$ or 1 mg/m^3.

Some knowledge of the natural composition of the earth's atmosphere is necessary in order to understand more fully the nature of the substances that are constantly being added through man's activities. We are concerned essentially with the lower atmosphere and whatever gaseous exchange takes place in the region up to about 40 000 feet [1] above the earth's surface. Above this layer, there is the stratosphere, which extends to about 60 miles, [2] and beyond this lies the ionosphere, where ultraviolet radiation from sunlight causes photochemical reactions that result in the formation of activated molecules and the splitting of molecules into atoms or ions.

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Cadle & Magill (1956) have indicated that the average composition of the lower atmosphere for those natural constituents that may be regarded as occurring in more or less constant concentration is substantially as shown in Table 1. To these constant components must be added a number of others of natural origin that may vary considerably in concentration. Water vapour in saturated air at 20°C is present to the extent of 17.118 g/m^3; consequently, in the range of about 25 %-100 % relative humidity, warm air may contain from approximately 5000 to over 20 000 p.p.m. of water vapour. In cold air the concentration may drop to one-tenth of this range.

TABLE 1

AVERAGE COMPOSITION OF THE ATMOSPHERE

Gas component	Composition (p.p.m., by volume)	Composition (p.p.m., by weight)
Nitrogen	780 900	755 100
Oxygen	209 500	231 500
Argon	9 300	12 800
Carbon dioxide	300	460
Neon	18	12.5
Helium	5.2	0.72
Methane	2.2	1.2
Krypton	1	2.9
Nitrous oxide	1	1.5
Hydrogen	0.5	0.03
Xenon	0.08	0.36

Source: Cadle & Magill (1956). (By permission from "Air pollution handbook," by P. L. Magill, F. R. Holden & C. Ackley, editors. Copyright 1956. McGraw Hill Book Company Inc.)

Other variable gaseous constituents of natural origin include oxides of nitrogen from electrical discharges during storms; sulfur dioxide, hydrogen fluoride and hydrogen chloride from volcanic disturbances; hydrogen sulfide from seepage of sour natural gas, from volcanoes or from action of sulfide bacteria; and ozone formed photochemically or by electrical discharge. Dust and aerosols of natural origin that are present in the atmosphere consist of salt particles from sea-water, various types of condensation nuclei, airborne particles from soil and vegetation, dust of meteoric origin, and bacteria spores and pollen. The concentrations of such substances, except in the immediate vicinity of natural sources, are extremely low, usually much less than one part per million for the gases and only a few micrograms per cubic metre for the particulates. Relatively high concentrations of ozone, between about 6 and 8 p.p.m., may exist in the stratosphere, but at ground level the range, over most areas where measurements are available, is from zero to about 5 parts per hundred million. However, ozone concentrations greater than 20 p.p.h.m. have been reported in Alaska by Wilson et al. (1952) and in southern California by Bartel & Temple (1952).

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[1] 1 ft = 0.3 m.

[2] 1 mile = 1.6 km.

Much has been learned about the transport and diffusion of matter and the manner in which the air is circulated by wind movement over great distances of the earth's surface by studies of some of these contaminants of natural origin. Sea-salt nuclei, originating from ocean breakers and salt spray along the coasts, have been found at distances inland of more than 1000 miles and at various heights up to 20 000 feet or more. Similarly, spores of fungi and bacteria have been collected with aircraft at distances of many hundreds of miles and allergens have been identified as far as 1500 miles from their most probable source. Micro-organisms of various types have been reported by Proctor (1934) at heights ranging up to 20 000 feet and at even higher altitudes—beyond 36 000 feet—by Rogers & Meier (1936).

Sources of Contaminants

The major sources, by far, of air contaminants from the activities of man are the products of combustion released in ever-increasing quantities through the use of fuels for domestic and industrial heating, power generation, transportation and other purposes. As a result of the strong upward trend in industrial production during the last decade, particularly in North America and certain parts of Europe, new air pollution problems have arisen whilst existing ones have been intensified. Apart from the emissions resulting from the combustion of fuels, important sources of contaminants exist in the atmospheric waste products of the iron and steel, non-ferrous metal smelting and refining, oil refining and petro-chemical, general inorganic and organic chemical, pulp and paper, and many other process industries.

It is of interest to compare the fuel consumption and estimated emissions of the principal classes of contaminants from the combustion products discharged to the atmosphere in the USA and Great Britain, as shown in Table 2. According to Rupp (1956) the amount of carbon dioxide emitted as a result of combustion of fuels in the USA in 1953 was at the rate of about 3000 million short tons [1] annually, and that of carbon monoxide about

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[1] 1 short ton = 0.9 metric ton. Throughout this chapter the unit " tons " refers to short tons.

TABLE 2

FUEL CONSUMPTION IN THE USA AND GREAT BRITAIN IN 1950 AND ESTIMATED EMISSIONS OF POLLUTANTS

Fuel	Fuel consumption per annum		Emission of pollutants per annum		
	USA	Great Britain	Contaminant	USA (millions short tons)[a]	Great Britain (millions short tons)[a]
Solid fuels (millions short tons)[a]			Smoke	5 *5*	2.2 *2.0*
Coal	498 *452*	226 *205*			
Lignite	3.1 *2.8*	negligible	Industrial dust and ash	7 *6*	0.9 *0.8*
Coke (except petroleum coke)	69 *63*	30 *27*			
Manufactured fuels	2.4 *2.2*	1.1 *1.0*			
Wood	[b]	0.5 *0.5*	Sulfur oxides as SO_2	19 *17*	5.8 *5.3*
Liquid fuels (millions bbls.)[c]					
Refined petroleum products, natural gasoline and benzol	2 110 *335*	123 *19.6*	Vapours: Hydrocarbons, nitrogen oxides, organic aldehydes and acids, ammonia, tars, etc.	42 *38*	[b]
Gaseous fuels (thousand millions cu. ft.)[d]					
Natural gas	5 200 *147*	negligible			
Manufactured gas	1 500 *42*	725 *20.5*			

Sources: Data on fuel consumption as given by Rupp (1956). Emission of pollutants — for USA: Rupp (1956); estimated for Great Britain from 1953 data in: Great Britain, Committee on Air Pollution (1955). (The data given by Rupp are reproduced by permission from "Air pollution handbook", by P. L. Magill, F. R. Holden & C. Ackley, editors. Copyright 1956. McGraw-Hill Book Company Inc.)

[a] The figures in italic type represent millions of metric tons.
[b] Not available (1954).
[c] The figures in italic type represent millions of cubic metres.
[d] The figures in italic type represent thousand millions of cubic metres.

60 million tons. Carbon dioxide is not normally considered to be an air contaminant and carbon monoxide becomes important only in special situations where the concentrations rise to sufficiently high levels, such as in

vehicular tunnels and at intersections of busy traffic arteries in congested city areas. Carbon monoxide in the outdoor atmosphere is present, usually, at levels far below the permissible threshold concentration of 100 p.p.m. for exposure during an 8-hour working day.

Within recent years increasing emphasis has been placed upon emissions of hydrocarbons, other organics and oxides of nitrogen by virtue of the possibility of photochemical interactions between such products after release to the atmosphere. In the past, the greatest concern has been exercised over the control of smoke, dust and sulfur dioxide emissions. However, waste exhaust products from the internal combustion engine are assuming prominence in atmospheric pollution studies in the light of experience in the Los Angeles area. Within North America the consumption of solid fuel has been decreasing and that of liquid and gaseous fuels has been rising steadily, especially in fuel usage for space- and water-heating. This trend has aided local abatement efforts in the control of smoke, dust and sulfur dioxide in a considerable number of urban areas.

Sulfur dioxide remains one of the major contaminants not only on an area-wide basis but also from specific, highly concentrated sources such as the metal smelting and oil refining industries and large coal-fired, electric power plants. It has been estimated by Katz & Cole (1950) that the annual emission of sulfur dioxide from a group of nickel-copper smelters in Canada has been as high as about 3 million tons during peak production years. From statistics available in some of the industrialized countries, the annual emission of sulfur dioxide from crude-oil refining is about 11 million tons, on the basis of an average content of 1.5 % sulfur in the crude. Most of this emission is within the USA. Other annual sulfur dioxide emission estimates for recent years of high production are as follows: copper smelters, about 11-12 million tons; lead and zinc smelters, 3.5-4 million tons; and from coal with an average sulfur content of 1.5 %, about 50-60 million tons.

The following examples will serve to illustrate the scale of emission that may occur from a single source.

Type of operation	*Daily capacity*	*SO_2 emission (tons per day)*
Copper smelter	2500 tons of concentrate, 30% S	1500
Oil refining	100 000 bbls., 1.5% S in crude	450
Coal-fired power plant	5000 tons of coal containing 3% S	300

Smelting operations, in particular, have been the cause of heavy damage to agricultural and forest areas and sulfur dioxide in the atmosphere has contributed to a major extent to the deterioration of materials such as metals, stone, cement, paper, paint, leather and textiles. In 1930, the emission of this gas from the stacks of the large lead-zinc smelter at Trail, British Columbia, reached an all-time high of about 20 000 tons per month, with the result that widespread damage to crops and forests occurred both in

Canada and in the adjacent part of the USA within the northern part of Stevens County, Washington State. The ensuing international litigation over this problem led to a pioneering effort on the part of the company involved to control this nuisance. Within the next decade a large new industry was created to convert the waste sulfur gases to sulfuric acid, ammonium sulfate, ammonium nitrate and phosphate fertilizer. Today, the Trail smelter recovers about 91 % of the sulfur dioxide, formerly wasted, by conversion into these valuable by-products. The Trail recovery plants have a capacity of 1300 tons of 100 % sulfuric acid and of 240 tons of synthetic ammonia per day, besides the necessary plant capacity for the production of the above fertilizers (Katz, 1952a; King, 1950).

Both in the USA and in Canada, there is an increasing trend to control atmospheric pollution in the smelting and oil refining industries through the recovery of waste sulfur dioxide and hydrogen sulfide by conversion to sulfuric acid or elemental sulfur. The latter by-product is usually the

TABLE 3
ESTIMATED RATES OF EMISSION OF CONTAMINANTS
FROM FUELS, INTERNAL COMBUSTION ENGINES AND INCINERATORS

Contaminant	Pounds of contaminants per short ton of fuel burned [a]						
	Coal	Oil	Gas	Engines		Incinerators	
				Automobile (gasoline)	Diesel (Diesel fuel)	Household	Municipal
Solids (carbon and particulates)	150 *75*	— —	— —	0.1 *0.05*	34 *17*	46.3 [b] *23.2* [b]	24 *12*
Sulfur oxides, as SO_2	80 *40*	60 *30*	— —	5.6 *2.8*	10 *5*	2 *1*	2 *1*
Nitrogen oxides, as NO_2	8 *4*	27 *13.5*	13.8 *6.9*	24.6 *12.3*	49 *24.5*	10.6 *5.3*	2 *1*
Ammonia	— —	— —	— —	0.6 *0.3*	— —	2.0 *1.0*	0.4 *0.2*
Acids, as CH_3 COOH	30 *15*	27 *13.5*	2.6 *1.3*	0.6 *0.3*	10 *5*	27.4 *13.7*	0.6 *0.3*
Aldehydes, as HCHO	— —	2.6 *1.3*	2.0 *1.0*	5.6 *2.8*	5 *2.5*	5.1 *2.6*	1.4 *0.7*
Other organics (incl. hydrocarbons)	20 *10*	9.2 *4.6*	2.8 *1.4*	141 *70.5*	[c]	274 *137*	1.2 *0.6*

Source: Stanford Research Institute (1950); and Larsen, Fischer & Hamming (1953); Magill & Benoliel (1952).

[a] The figures in italic type represent kilograms of contaminants per metric ton of fuel burned.

[b] Ether-soluble and ether-insoluble aerosols.

[c] Not available.

favoured one where hydrogen sulfide from oil refining or processing of sour natural gas is concerned. In Great Britain a considerable amount of by-product sulfuric acid is produced from zinc concentrates and spent oxide.

Some comparative data on the amounts of contaminants that may be discharged to the atmosphere from the combustion of coal, oil, gas, gasoline, and rubbish in fuel-burning equipment, automobiles, Diesel engines and incinerators are shown in Table 3. The combustion of coal results in the highest emissions of solids, largely fly ash and soot, sulfur oxides and acids. Exhaust gases from automobiles contribute large quantities of unburned hydrocarbons and other organics and considerable amounts of nitrogen oxides. Diesel engines emit the highest percentage of nitrogen oxides, with oil-burning installations next in order of rank. The burning of rubbish and paper in domestic incinerators yields relatively large quantities of organics and other contaminants in comparison with the emissions from the much more efficient, properly designed, municipal incinerators.

The emission quantities cited in Table 3 are only rough, approximately average figures that would vary with the composition of the fuel and operating characteristics of the equipment or engines in particular cases. For example, great variations may occur in the combustion products and contaminants from automobiles under various driving conditions and also in relation to age of the engine in the vehicle. As a rule there are greater quantities of contaminants emitted from old cars than from new cars. Unburned hydrocarbons are discharged in much greater quantities during deceleration than in acceleration or steady driving. With the engine idling, the emission of hydrocarbons is greater than that during steady driving or acceleration but is still considerably less than the emission during deceleration.

Rupp (1956) has attempted to summarize the total emissions of some of the more widely prevalent organic vapour and other contaminants within the USA. On an annual basis, emissions of organic compounds in combustion products of coal, oil, gasoline and refuse, etc., are estimated at 21 million tons; evaporation products from losses of hydrocarbons, natural gas, solvent vapours and other materials prior to combustion yield 11 million tons of additional organics. Nitrogen compounds expressed as nitrogen dioxide amount to 8 million tons and halogen compounds to about 2 million tons.

Los Angeles County Emissions

The Los Angeles district suffers from recurrent haze and smog accompanied by irritation of the eyes, nose and throat of large numbers of inhabitants, and considerable economic losses in damage to certain crops. Other effects are reduced visibility, objectionable odour and rapid deterioration of rubber. The phenomenal growth of population and industry in the area since the Second World War and the limitations on natural air cleaning processes imposed by meteorological and topographical factors have resulted in an air pollution problem of unusual complexity. This region now has a population of about 5 million and is the largest subtropical, heavily industrialized, urban area in the world. Although smog was known in Los Angeles long before the Second World War, the rapid expansion of industrial and public activities during and after the war accentuated the air pollution problem. The more noticeable smog visitations occur on about 60 days of the year and the annual average of temperature inversions has been estimated at about 262 days.

The topography of this county is a large basin fronting the Pacific Ocean to the west, with a ring of high mountains on the other three sides. An inversion layer of stratified air extends over this basin at levels which usually range between 1000 and 3000 feet. As a rule, the inversion layer has an upward slope both to the west and to the east. Because it rests against the mountain range on the east side of the basin, the flow of air eastward, out of the area, is prevented. The sea breeze in the daytime and the land breeze at night merely shift the inversion layer up and down but do not disperse it.

This temperature inversion layer is brought about by the presence of a semi-permanent high pressure area over the North Pacific Ocean, extending from the west coast of the USA to beyond the Hawaiian Islands. This high pressure area is present during the major portion of the summer and autumn months, although its position may vary in longitude and latitude. It is an enormous, elliptical column of air, moving in a clockwise direction on its axis, and inclined slightly to the south-west. The air paths at intermediate levels in this column are steeply inclined to the earth's surface, the lower edge of this layer being adjacent to the California coast and its upper edge being suspended in mid-Pacific to the north of the Hawaiian Islands. The air moving towards California around the northern side of this pressure area is descending, moving through levels of increased pressure, and is therefore being warmed by compression. In consequence, the air arriving over Los Angeles County from the ocean, at levels above the surface air layer, is already much warmer than the surface air. This temperature difference is accentuated when air over the ocean surface, which has been cooled from below by contact with relatively cold ocean water off-shore, is swept inland into the basin (Beer & Leopold, 1947).

The inversion layer is like a canopy over the Los Angeles basin, preventing both vertical and lateral dispersion of contaminants. The natural haze, composed of salt from ocean spray and dust particles from soil and vegetation, is augmented by smoke, fumes, and gases from industrial and domestic activities, including vehicular traffic. The larger aerosol particles are removed by settling, but the fine aerosols, and gaseous pollutants tend to build up at the top of the atmospheric layer which lies just below the base of the inversion stratum. The worst conditions occur when the man-made

TABLE 4

ESTIMATED EMISSIONS OF AEROSOLS, ACID GASES, AND ALDEHYDES TO THE ATMOSPHERE IN SHORT TONS PER DAY, LOS ANGELES COUNTY, 1940, 1948 AND 1951 *

Source	Aerosols *a*			Oxides of nitrogen *b*			Sulfur dioxide			Aldehydes *c*			Organic acids *d*		
	1940	1948	1951	1940	1948	1951	1940	1948	1951	1940	1948	1951	1940	1948	1951
Total	220 *200*	350 *318*	225 *205*	160 *145*	260 *235*	255 *231*	385 *350*	595 *540*	300 *272*	25 *22*	35 *32*	25 *23*	100 *91*	140 *127*	90 *81*
Chemical, paint, roofing rubber, soap, food, fertilizer, metal and mineral industries	55 *50*	104 *94*	61 *55*	*e*	*e*	*e*	11 *10*	23 *21*	27 *24*	*e*	*e*	*e*	*e*	*e*	*e*
Fuel oil burning *f*	3 *3*	5 *5*	3 *3*	50 *45*	85 *77*	85 *77*	100 *91*	170 *154*	170 *154*	*e*	*e*	*e*	3 *3*	7 *6*	7 *6*
Transportation — Diesel	3 *3*	5 *5*	7 *6*	4 *4*	7 *6*	10 *9*	1 *1*	2 *2*	2 *2*	— —	1 *1*	1 *1*	1 *1*	2 *2*	3 *3*
Transportation — gasoline	14 *13*	23 *21*	25 *23*	71 *64*	120 *109*	132 *120*	13 *12*	20 *18*	21 *19*	7 *6*	10 *9*	10 *9*	4 *4*	7 *6*	7 *6*
Petroleum refining	— —	8 *7*	5 *5*	— —	— —	— —	260 *236*	380 *345*	80 *73*	— —	— —	— —	— —	— —	— —
Incineration — domestic	60 *54*	90 *82*	90 *82*	15 *14*	21 *19*	21 *19*	*e*	*e*	*e*	8 *7*	10 *9*	10 *9*	42 *38*	55 *50*	55 *50*
Incineration — commercial, industrial, municipal and dumps	85 *77*	115 *104*	34 *31*	20 *18*	27 *24*	7 *6*	*e*	*e*	*e*	10 *9*	14 *13*	4 *4*	50 *45*	69 *63*	18 *16*

Source: Los Angeles County Air Pollution Control District (1951), Table XX, p. 39. (By permission from the Los Angeles County Air Pollution Control District.)

* The figures in italic type represent metric tons per day.

a Includes smoke, dusts, fumes and mists. *b* Reported as nitrogen dioxide. *c* Reported as formaldehyde.

d Reported as acetic acid. *e* Less than one ton.

f Includes fuel oil burned in all categories except transportation. All values are based on summer fuel oil usage, which is approximately one-half the winter usage.

TABLE 5

ESTIMATED EMISSIONS OF HYDROCARBONS TO THE ATMOSPHERE IN SHORT TONS PER DAY, LOS ANGELES COUNTY, 1940, 1948 AND 1951 *

Source	Total hydrocarbons			Olefinic unsaturation						Saturates [b] more than 3 carbon atoms per molecule			Acetylene		
				Total			4, 5 and 6 carbon atoms per molecule [a, d]								
	1940	1948	1951	1940	1948	1951	1940	1948	1951	1940	1948	1951	1940	1948	1951
Total	2 780 *2 523*	2 090 *1 896*	2 130 *1 933*	170 *155*	285 *259*	300 *271*	60 *54*	105 *96*	115 *105*	880 *799*	1 370 *1 243*	1 420 *1 288*	50 *45*	85 *77*	90 *82*
Chemical, paint, roofing rubber and soap industries	13 *12*	25 *23*	35 *32*	c	c	7 *6*	c	c	4 *4*	13 *12*	25 *23*	28 *25*	c	c	c
Petroleum marketing	67 *61*	115 *104*	120 *109*	10 *9*	17 *15*	18 *16*	8 *7*	16 *15*	17 *15*	57 *52*	95 *86*	102 *93*	— —	— —	— —
Petroleum production[e]	1 700 *1 542*	270 *245*	270 *245*	c	c	c	c	c	c	120 *109*	100 *91*	100 *91*	c	c	c
Petroleum refining[f]	500 *454*	830 *753*	830 *753*	80 *73*	135 *122*	135 *122*	27 *24*	47 *43*	48 *44*	370 *336*	610 *553*	610 *553*	c	c	c
Transportation — gasoline[g]	500 *454*	850 *771*	875 *794*	80 *73*	13 *12*	140 *127*	25 *23*	42 *38*	46 *42*	320 *290*	540 *490*	580 *526*	50 *45*	85 *77*	90 *82*
	1950			1950			1950			1950			1950		
Automobile exhaust [h]	850 *771*			372 *337*			143 *130*			221 *200*			33 *30*		

Source: Los Angeles County Air Pollution Control District (1951), Table XXI; Stanford Research Institute (1954), Table XXXIX; and Southern California Air Pollution Foundation (1954), pp. 11-19.

* The figures in italic type represent metric tons per day.

[a] Some authorities consider the oxidation products of these compounds to be a major factor in air pollution. [b] Includes aromatics. [c] Less than one ton.

[d] Other emissions, principally methane and ethane: 1948—92 tons (83 metric tons); 1950—213 tons (193 metric tons).

[e] Includes losses from production and field operations for crude oil and natural gas. The 1940 figure includes release of large quantities of natural gas directly to the atmosphere.

[f] Includes evaporation losses from gasoline and crude oil storage at refineries.

[g] Includes breathing losses from gasoline tanks and carburettors.

[h] Based upon research work as set forth in Stanford Research Institute (1954), pp. 115-126, and as developed in Southern California Air Pollution Foundation (1954), 11-19. Figures do not include breathing losses from gasoline tanks and carburettors, which are estimated at 1 % of gasoline consumption (Southern California Air

POLLUTANTS EMITTED TO THE ATMOSPHERE BY COMBUSTION PROCESSES IN SHORT TONS PER DAY, LOS ANGELES COUNTY, 1950 AND 1953 *

Material burned and source	Quantity burned		Emissions											
			Aldehydes (as HCHO)		Ammonia		Nitrogen oxides (as NO_2)		Sulfur oxides as (SO_2)		Acids * (as CH_3COOH)		Organics	
	1950	1953	1950	1953	1950	1953	1950	1953	1950	1953	1950	1953	1950	1953
Total			71.5 *65.5*	85.5 *77.0*	12.1 *10.6*	14.1 *12.6*	384.5 *349.0*	463 *420*	344.2 *312.3*	411.2 *373.2*	134.5 *122.0*	156.6 *142.5*	1 267 *1 150*	1 534 *1 392*
Fuel gas, total	20 000 *18 140*	23 760 *21 560*	20.0 *18.0*	23.5 *21.5*	4.0 *3.5*	5.0 *4.5*	137.0 *124.0*	164 *149*	83.0 *75.5*	103.0 *93.5*	26.0 *23.5*	30.0 *27.5*	30 *27*	35 *32*
General public	6 680 *6 050*	7 670 *6 960*	6.5 *6.0*	7.5 *7.0*	— —	— —	46.0 *41.5*	53 *48*	— —	— —	9.0 *8.0*	10.0 *9.0*	10 *9*	11 *10*
Petroleum industry	8 720 *7 910*	10 800 *9 800*	9.0 *8.0*	11.0 *10.0*	4.0 *3.5*	5.0 *4.5*	60.0 *54.5*	75 *68*	83.0 *75.5*	103.0 *93.5*	11.0 *10.0*	13.0 *12.0*	13 *12*	16 *15*
Other industry	4 600 *4 170*	5 290 *4 800*	4.5 *4.0*	5.0 *4.5*	— —	— —	31.0 *28.0*	36 *33*	— —	— —	6.0 *5.5*	7.0 *6.5*	7 *6*	8 *7*
Fuel oil, total	7 500 *6 810*	8 830 *8 010*	9.0 *8.5*	10.0 *8.5*	— —	— —	102.0 *92.5*	119 *107*	227.0 *205.5*	266.0 *241.0*	103.0 *93.5*	120.0 *109.0*	35 *32*	40 *36*
General public	2 060 *1 870*	2 370 *2 150*	2.5 *2.5*	3.0 *2.5*	— —	— —	28.0 *25.5*	32 *29*	62.0 *56.0*	71.0 *64.5*	28.0 *25.5*	32.0 *29.0*	10 *9*	11 *10*
Petroleum industry	2 270 *2 060*	2 820 *2 560*	2.5 *2.5*	3.0 *2.5*	— —	— —	31.0 *28.0*	38 *34*	68.0 *61.5*	84.0 *76.0*	31.0 *28.0*	38.0 *34.5*	10 *9*	12 *11*
Other industry	3 170 *2 880*	3 640 *3 300*	4.0 *3.5*	4.0 *3.5*	— —	— —	43.0 *39.0*	49 *44*	97.0 *88.0*	111.0 *100.5*	44.0 *40.0*	50.0 *45.5*	15 *14*	17 *15*
Refuse, total	7 900 *7 170*	7 560 *6 860*	10.5 *10.0*	12.0 *10.5*	4.1 *3.6*	4.1 *3.6*	2.5 *2.5*	3 *3*	2.2 *2.2*	2.2 *2.2*	2.5 *2.5*	2.6 *2.5*	385 *350*	443 *402*
General public	4 400 *3 990*	5 060 *4 590*	8.0 *7.5*	9.0 *8.0*	4.0 *3.5*	4.0 *3.5*	1.0 *1.0*	1 *1*	2.0 *2.0*	2.0 *2.0*	2.0 *2.0*	2.0 *2.0*	360 *327*	414 *376*
Industry	3 500 *3 180*	2 500 *2 270*	2.5 *2.5*	3.0 *2.5*	0.1 *0.1*	0.1 *0.1*	1.5 *1.5*	2 *2*	0.2 *0.2*	0.2 *0.2*	0.5 *0.5*	0.6 *0.5*	25 *23*	29 *26*
Gasoline	1 550 *1 410*	14 380 *13 050*	32.0 *29.0*	40.0 *36.5*	4.0 *3.5*	5.0 *4.5*	143.0 *130.0*	177 *161*	32.0 *29.0*	40.0 *36.5*	3.0 *2.5*	4.0 *3.5*	817 *741*	1 016 *922*

Source: Stanford Research Institute (1954), Table XLVI, p. 133. (By permission from the Western Oil and Gas Association, Los Angeles.)

* The figures in italic type represent metric tons per day.

Note: Fractions of ton per day are estimated to the nearest 0.5 ton per day except for quantities of less than 1.0 ton.

TABLE 7
TOTAL EMISSIONS OF AIR CONTAMINANTS TO THE ATMOSPHERE IN SHORT TONS PER DAY, LOS ANGELES COUNTY, JULY 1958 *

ORGANIC GASES		AEROSOLS	INORGANIC GASES	
Hydrocarbons	1 450 *1 320*	100 *90*	Oxides of nitrogen	700 *635*
Aldehydes and ketones	65 *60*		Sulfur dioxide	550 *495*
Other	140 *125*		Carbon monoxide	5 600 *5 070*
			Other	5 *5*
Total	1 655 *1 505*	100 *90*	Total	6 855 *6 205*

EMISSIONS OF AIR CONTAMINANTS TO THE ATMOSPHERE BY SOURCE

Source	Organic gases				Inorganic gases			
	Hydro-carbons	Aldehydes and ketones	Other organics	Aerosols	Nitrogen oxides	Sulfur dioxide	Carbon monoxide	Other inorganics
Transportation								
Automobile [a]	750 *680*	9 *8*	11 *10*	23 *21*	320 *290*	35 *32*	3 200 *2 903*	[b]
Trucks and buses (gasoline)	250 *227*	3 *3*	4 *4*	9 *8*	110 *100*	13 *12*	1 000 *907*	[b]
Trucks and buses (Diesel)	[b]	[b]	1 *1*	2 *2*	3 *3*	1 *1*	2 *2*	[b]
Petroleum								
Refining	55 *50*	2 *2*	[b]	5 *5*	5 *5*	50 *45*	1 200 *1 089*	4 *4*
Storage [c]	80 *73*	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*
Marketing	55 *50*	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*
Production	60	[b]	0	0	[d]	[b]	[d]	0 *0*

Surface coating	90 *82*	21 *19*	20 *18*	2 *2*	0 *0*	0 *0*	0 *0*	0 *0*
Degreasing	20 *18*	1 *1*	45 *41*	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*
Dry cleaning	20 *18*	0 *0*	8 *7*	*b*	0 *0*	0 *0*	0 *0*	0 *0*
Other	50 *45*	20 *18*	25 *23*	1 *1*	0 *0*	0 *0*	0 *0*	0 *0*
Combustion of fuels								
Liquid	1 *1*	1 *1*	4 *4*	30 *27*	160 *145*	350 *318*	*b*	*b*
Gaseous	5 *5*	1 *1*	3 *3*	8 *7*	100 *91*	50 *45*	1 *1*	*b*
Incineration								
Industrial	*b*	*b*	1 *1*	1 *1*	*b*	*b*	1 *1*	*b*
Commercial	*b*	0 *0*	*b*	*b*	0 *0*	0 *0*	*b*	*b*
Municipal	*b*	0 *0*	*b*	*b*	*b*	0 *0*	*b*	*b*
Other	*b*	*b*	*b*	*b*	0 *0*	0 *0*	2 *2*	0 *0*
Chemical								
Sulfur and sulfuric acid	0 *0*	0 *0*	0 *0*	0 *0*	0 *0*	45 *41*	0 *0*	0 *0*
Other	16 *15*	8 *7*	16 *15*	3 *3*	*d*	1 *1*	*d*	1 *1*
Metals	0 *0*	0 *0*	0 *0*	6 *5*	0 *0*	*b*	180 *163*	0 *0*
Minerals	2 *2*	0 *0*	0 *0*	6 *5*	0 *0*	*b*	0 *0*	0 *0*
Miscellaneous	*b*	*b*	*b*	2 *2*	*b*	*b*	*d*	*d*

Source: Total Pollution Card, July 1958. (By permission from Los Angeles County Air Pollution Control District, Engineering Division.)

* The figures in italic type represent metric tons per day. *a* Includes trucks under 4000 lb. (1800 kg) unladen weight. *b* Negligible—less than 0.5 ton per day.
c Includes storage of petroleum products at refining and marketing installations. *d* Unknown.

pollution accumulates over several days in calm weather and the base of the inversion layer is forced to its lowest level, thus bringing the polluted air to the ground (Stanford Research Institute, 1948-50).

The major contributors to air pollution in the Los Angeles area are motor vehicles, combustion of fuel oil and gas, petroleum refining, vapour losses from gasoline marketing, distribution, and storage, and the incineration of refuse. Several organizations have presented estimates of the emissions of various types of contaminant released to the atmosphere as a result of industrial and public activities. These emissions are shown in Tables 4-7. There are about 2.5 million motor vehicles in Los Angeles County, or about one vehicle for every two persons. From this source alone about 1200 tons of hydrocarbons (mainly gasoline vapours) are discharged daily into the atmosphere. In 1953 it was estimated by the Stanford Research Institute that the combustion processes of all types released to the air about 1500 tons of organics daily. The practice of burning combustible rubbish in household incinerators has helped to accentuate the troublesome smog problem. About 1.5 million back-yard incinerators burned an average of more than 5000 tons of refuse per day in 1955, but this source of inefficient combustion has recently been brought under control by regulation under the County Air Pollution Control District. Other important contaminants in this area, besides hydrocarbons, are oxides of nitrogen, sulfur oxides, acids, aldehydes and aerosols.

The emissions of smoke, dust, fumes and sulfur oxides are relatively low for the huge scale of industrial and public activities in this area. A major factor has been the strict enforcement of county air pollution control regulations, which limit the discharge of dust on a process weight basis up to a top limit of 40 lb.[1] per hour for any one industrial operation (McCabe et al., 1949). To meet such provisions, high efficiency dust collection equipment must be installed on all plants that may emit fly ash, metal and mineral dusts, chemical fumes and mists. Also prohibited is the discharge of smoke of No. 2 Ringelmann density or greater, and of sulfur dioxide in stack gases to an amount greater than 0.2 % by volume.

Emissions in the Greater Detroit-Windsor Area

In 1948, officials of the cities of Detroit, Mich. and Windsor, Ont. requested the US State Department and the Canadian Department of External Affairs, respectively, to assist in solving a problem which apparently could not be solved by the two municipal governments nor the state or provincial governments concerned. The problem was to control smoke being emitted by vessels plying the international Detroit River. The US Department of State and the Canadian Department of External Affairs, under the terms of the Boundary Waters Treaty of 1909 between the two countries, jointly referred the problem to the International Joint Commission. The reference, jointly agreed to in January 1949, requested the Commission to determine whether or not the air over and in the vicinity of Detroit and Windsor was polluted to an extent detrimental to the area and whether or not ships plying the river were contributing to the pollution and what other sources were responsible. If the vessels were found to be responsible for detrimental air pollution, the Commission was requested to recommend remedial measures, to indicate what would be the cost of the corrective measures and by whom the costs should be borne.

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[1] 1 lb. = 0.45 kg.

To obtain information necessary to reply to the reference, the US and Canadian Sections of the International Joint Commission appointed a joint Technical Advisory Board. This Board, with great assistance from many agencies and individuals, conducted the required investigations and presented a final report (1959). The results with respect to sources and emissions of contaminants provide some interesting contrasts to those of the Los Angeles area.

The Detroit Metropolitan Area is the third largest manufacturing centre in the USA, although it is only the fifth largest in population. Its manufacturing employment is exceeded only by New York and Chicago. Approximately 3.25 million people live in the international metropolitan area. The extensive industrialization, dominated by the automobile and primary metals industries, together with the extensive use of coal and other fuels, use of vehicular fuels, burning of refuse, and a wide assortment of other activities, gives rise to a large atmospheric pollution load.

Generally speaking, annual fuel usage consists of about 13 million tons of coal, 4.3 million tons of fuel oils, and 3.8 million tons of natural gas. Daily use of gasoline is about 3.4 million gallons [1] and of Diesel fuel, 42000 gallons. An estimated 1235 tons of refuse per day are burned in municipal incinerators, 1300 tons in domestic incinerators, and 400 tons in commercial and industrial incinerators.

Blast furnaces and steel mills comprise a significant proportion of the manufacturing activity in the area. About 7 million tons of pig iron and steel are produced each year. There are also about 30 iron and steel foundries producing about 4300 tons of castings per day. Approximately 100 establishments are engaged in non-ferrous founding and secondary metal refining. About 40 % of all manufacturing workers are employed in plants producing automobiles, trucks, and buses, and parts for these vehicles. The chemical industry comprises about 200 plants and there is some petroleum refining. A host of other industrial activities also contribute to the pollution of the atmosphere. Solvent losses to the atmosphere from use of

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[1] 1 US gallon = 3.8 l.

TABLE 8

ESTIMATED PRINCIPAL AIR POLLUTION EMISSIONS IN SHORT TONS PER DAY, GREATER DETROIT-WINDSOR AREA [*] [†]

Source of pollution	General public				Industry and commerce			
	Solids	Sulfur oxides (as SO_2)	Nitrogen oxides (as NO_2)	Organics *a*	Solids	Sulfur oxides (as SO_2)	Nitrogen oxides (as NO_2)	Organics *a*
Use of gas	*b*	1	48	25	*b*	1	26	15
		1	*44*	*23*		*1*	*24*	*14*
Use of coal	210	283	42	263	193	408	44	276
	191	*257*	*38*	*239*	*175*	*370*	*40*	*250*
Use of petroleum	1	126	55	89	2	230	98	158
	1	*114*	*50*	*81*	*2*	*209*	*89*	*143*
Automotive fuel use	1	26	111	597	<1	4	19	99
	1	*24*	*101*	*542*	*<1*	*4*	*17*	*90*
Gasoline handling	*b*	*b*	*b*	129	*b*	*b*	*b*	21
				117				*19*
Refuse burning	18	2	1	162	5	1	1	41
	16	*2*	*1*	*147*	*5*	*1*	*1*	*37*
Solvent usage	*b*	*b*	*b*	78	*b*	*b*	*b*	105
				71				*95*
Metallurgical industry	—	—	—	—	743	*c*	*d*	*d*
	—	—	—	—	*674*			
Petroleum refining	—	—	—	—	1	10	*d*	150
	—	—	—	—	*1*	*9*		*136*
Total	230	438	257	1 343	945	654	188	865
	209	*398*	*234*	*1 220*	*858*	*594*	*171*	*784*

[*] Does not include carbon monoxide or carbon dioxide.
[†] The figures in italic type represent metric tons per day.

a Includes gasoline and solvents.
b Nil or negligible.
c Included in " Use of coal ".
d Data not available.

paints, dry cleaning, plastics moulding, gluing, degreasing, etc., constitute a sizable atmospheric pollution load.

Vessels plying the Detroit River contribute to the pollution of the atmosphere by emissions of black smoke, fly ash, and gaseous contaminants from the combustion of coal and oil. The proportion of the total pollution emissions in the area attributable to vessel operations is quite small. However, the emissions from vessels are concentrated on the river and are particularly objectionable because of the close proximity of residential, recreational, and civic land uses at many points along the river. Since 1952, a major reduction in pollution from this source has been effected through the operation of a voluntary control programme and conversions to more efficient fuel-burning installations.

An approximate inventory of the principal pollution emissions to the atmosphere from all of man's activities in the area is shown in Table 8. Emissions to the atmosphere in the Canadian portion of the area were found to be about 10 % of those in the United States portion. In all, about 4940 tons of contaminants are discharged daily, excluding carbon monoxide or carbon dioxide. Owing to the large-scale consumption of solid fuel, the emissions of particulates and sulfur oxides are far greater than those estimated for the Los Angeles area, where virtually no coal is used.

Both meteorological and topographical factors are, in general, favourable to the natural dispersion and dilution of these relatively large amounts of contaminants. For the most part the land in the Detroit River area is predominantly flat and rises only slightly at some distance from the river. The area lies in the mid-latitude belt of a general west-to-east flow of air during all seasons of the year. The general features of the circulation bring a succession of changing pressure systems and air masses across the area, and the topography does not favour any localized areas of stagnating air. Conditions favourable for the accumulation of air contaminants near the ground can develop, principally, during night hours in the warmer portion of the year, but such inversions are usually dispersed some hours after sunrise. Prolonged inversions occur only at infrequent intervals with accompanying increases in the average concentration levels of contaminants.

Nature and Properties of Pollutants

Atmospheric contaminants may be classified simply into two types:

(1) Airborne particulates consisting of solid and liquid particles of a wide range of sizes varying from relatively large particles over 20 μ in diameter to aerosols or suspensions of fine particles extending into the submicron region, down to less than 0.05 μ.

(2) Gases or vapours, including the permanent gases and those compounds that have boiling points below about 200°C.

Although upwards of about 100 specific substances have been identified as contaminants arising from the varied activities of man, there are many more products that are still unknown. The particulate portion contains more than twenty metallic elements that may be determined by spectrographic analysis of the inorganic fraction. The organic fraction of particulates is even more complex and contains large numbers of aliphatic and aromatic hydrocarbons, acids, bases, phenols, and many other types of compound. Furthermore, new products are known to be created by the interaction of contaminants with each other after discharge to the atmosphere.

These mechanisms include photochemical and free radical reactions, oxidation and reduction, polymerization, condensation, catalysis and others.

Dust or aerosol particles may show greatly enhanced surface activity by virtue of increased surface area, adsorption of gas molecules or other properties that facilitate chemical reactions. Many substances that oxidize only slowly in their massive state will oxidize with extreme rapidity or explode when dispersed as fine dust in air. Adsorption and catalytic phenomena are influenced particularly by the state of dispersion of the solid or liquid phases in the gas dispersion medium of heterogeneous systems. Thermal radiation effects become much more pronounced if suspended solid or liquid particles are present in the air. Such particles absorb radiation and conduct heat rapidly to the surrounding gas molecules, which may be quite transparent to the radiant energy. Aerosol particles may act as condensation nuclei to facilitate the condensation of water vapour upon them and thus promote the formation of fog or ground mists.

Aerosols

Particles dispersed in a gaseous medium are collectively termed an aerosol. The terms smoke, fog, haze and dust are used to describe particular types of aerosol, depending on the size, shape and characteristic behaviour of the dispersed particles. The particles themselves will be spherical if liquid, but will have various shapes if solid—although solid particles tend to behave like spheres in some aspects.

Aerosols may be formed by two general methods: (*a*) condensation processes in which clusters of molecules come together to produce particles of colloidal dimensions, and (*b*) dispersion processes in which coarse matter is further split up into fine particles (Green & Lane, 1957).

A familiar example of the condensation process is the formation of a cloud of liquid droplets when warm moist air rises into the cooler upper atmosphere. The heating of volatile substances causes them to evolve vapours, which, on cooling, form small liquid and solid particles. Fine dust particles, nucleating agents, ions existing in the vapour or carrier gas, polar molecules such as sulphuric acid, or molecular aggregates of the vapour itself, may form the nuclei upon which the vapour will condense to produce the aerosol.

Some examples of mechanical dispersion processes are the production of fly ash in furnaces burning pulverized fuel, the emission to the atmosphere of solid by-products from foundries and size reduction operations, the solids discharged during " slips " from blast furnaces, the dust discharged in catalytic cracking operations, as in the petroleum industry, and the particulates dispersed by the movement of vehicles on city streets.

TABLE 9
PROPERTIES OF SOME TYPICAL AEROSOLS

Type of dispersal system	Size range of particles diameter (μ)	Electro-magnetic spectrum (μ)	Terminal velocity (cm/sec.) due to gravity settling in air, 20° C and 1 atmosphere
Raindrops	5 000-500		Turbulent motion $v_t = k_1$ pD for particles down to about 1000 μ
Natural mist	500-40		p = particle density D = particle diameter (μ)
Natural fog	40-1.0		
		DUSTS	
Foundry sand	2 000-200		
Ground limestone, fertilizers	800-30		Intermediate region for particles between 1000-100 μ.
Sand tailings from flotation	400-20		$v_i = k_2 p^{2/3} D$
Pulverized coal	400-10		Stokes' law for streamline region, applicable to particles from 100-1.0 μ
Ground sulphide ore for flotation	200-4	Infra-red 420-0.7 μ	$v_s = K\, pD^2/n$
Foundry shake-out dust	200-1		
Cement	150-10		n = coefficient of viscosity of air, poises
Fly ash	80-3		$v_s = k_3 pD^2$
Silica dust in silicosis	10-0.5		
Pigments	8-1.0		
Pollens	60-20		
Plant spores	30-10		
Bacteria	15-1.0		
		FUMES AND MISTS	
Metallurgical fumes	100-0.1		Cunningham's correction for particles in range 1.0-0.10 μ
H_2SO_4 concentrator mist	10-1.0	Visible spectrum 0.7-0.40 μ	
Alkali fume	2-0.1		$v_c = v_s \left(\frac{1 + 1.72\,l}{D}\right)$
SO_3 mist	3-0.5		l = mean free path of gas molecules (μ)
NH_4Cl fume	2-0.1		
Zinc oxide fume	0.3-0.03		$v_c = v_s \left(\frac{1 + 0.172}{D}\right)$ in air

Source: Miller (1938) and Silverman (1951).

TABLE 9

PROPERTIES OF SOME TYPICAL AEROSOLS *(continued)*

Type of dispersal system	Size range of particles diameter (μ)	Electro-magnetic spectrum (μ)	Terminal velocity (cm sec.) due to gravity settling in air, 20° C and 1 atmosphere
		SMOKES	
Oil smoke	1.0-0.03		Velocity due to Brownian Motion exceeds velocity of gravity settling for particles less than 0.1 μ
Rosin smoke	1.0-0.01		
Tobacco smoke	0.15-0.01	Ultraviolet ↕ 0.40-0.015 μ	Einstein equation
Carbon smoke	0.2-0.01		$x = k_4 \quad t/D$
Normal impurities in quiet atmosphere	1.0-0.01		x = average displacement in cm of spherical particle in air in time, t sec.
			Spheres — Irregular shape $k_1 = 24$ — 16 $k_2 = 0.41$ — 0.26 $k_3 = 0.0030$ — 0.002 $k_4 = 0.00068$ — —

Source: Miller (1938) and Silverman (1951).

Particle Size

The particulates in the atmosphere are usually classified according to particle size into deposited matter (dustfall) and finely divided, suspended matter (aerosols). The fly ash, coarse dust, and dirt which consists of particles larger than about 10 μ in diameter settle out of the air fairly rapidly and constitute the major portion of the deposited matter. The finer particles, ranging from about 5 μ down to 0.1 μ or less, form suspensions in air which are mechanically stable. The smaller particles in this range exhibit Brownian movement and may be dispersed in the atmosphere by wind currents almost like a gas (Katz & Clayton, 1953).

Aerosols generally include, by definition, all solid and liquid particles in the diameter range from about 100 μ down to 0.01 μ. The physical properties of some typical aerosols in relation to the electro-magnetic spectrum are shown in Table 9.

Fog, mist or cloud consists of liquid particles which are mainly water if they are of natural origin. Fume is formed by high temperature volatilization or by chemical reaction. Dust is a general term usually applied to solid particles only, such as those generated from mechanical operations and natural processes that involve dispersion and disintegration of material. Smoke is produced during combustion or destructive distillation and may contain either solid or liquid particles.

In the Detroit-Windsor area, studies on the particle size-distribution of air-borne particulates showed that 87 % of samples collected with a standard impinger and 83.9 % of samples collected with a thermal precipitator were less than 2 μ in size. About 99 % of the particles collected were below 10 μ in diameter (Katz & Clayton, 1953). In the London area Billington & Saunders (1947) measured the size-distribution of particles in smoke and found that the geometric mean diameter of the particles was in the range 0.5-1.1 μ. Cartwright, Nagelschmidt & Skidmore (1956) report similar figures for samples collected from the air in the Sheffield area.

The size of aerosols has an important bearing on the penetration of particles beyond the respiratory passages into the lungs. The coarser material is excluded by the nasal passages so that few particles may pass if they are larger than about 5 μ in diameter. Below 5 μ the penetration into the lungs increases rapidly, but when the size becomes less than about 1 μ the retention in the lung begins to decrease. Very small particles (less than 0.5 μ) penetrate readily but only a small fraction is retained in the lung (Johnstone, 1952).

Particle Number Concentration

The number concentration of suspended particles per unit volume in a polluted atmosphere may be extremely large, although the mass concentration may be low in comparison with that of gas contaminants. Thus, for example, an aerosol concentration of 0.1 mg/m^3, assuming unit density and 0.5 μ particle diameter, would correspond to 45 million particles per cubic foot.[1] Even in relatively clean air, aerosol particles numbering several million per cubic foot may be present (Katz & Clayton, 1953). In extreme smog conditions 2×10^9 or 3×10^9 particles per cubic foot may be reached. The concentration of particles greater than 0.5 μ in diameter in Los Angeles smog is estimated at about 56 million per cubic foot.

Coagulation

In the examination of submicron dust the electron microscope is widely used. However its usefulness has been limited by the difficulty of identifying particles by their external morphology, which is all that this instrument reveals. It does, nevertheless, provide valuable information on the size, shape and extent of coagulation of particles of submicron size, beyond the range of the optical microscope. Quite often some estimate of the relative abundance and state of aggregation of various particles can be obtained. Photomicrographs of carbon particles prepared from samples collected on millipore filters (Type AA, Lovell Chemical Company) during haze or smog periods in Windsor, show rather characteristic but similarly formed filaments of carbon consisting of aggregates of particles of 0.05 μ and 0.2 μ size range. Each filament, however, was composed of particles of similar size only (Shore & Katz, 1954). This process of filament formation by coagulation resembles a bimolecular mechanism, with kinetics similar to a second order reaction (Rodebush, 1950).

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[1] 1 cu.ft. = 0.028 m^3.

The rate at which coagulation occurs depends on the concentration and is almost independent of particle size. The time, t, in seconds to reduce the number of particles in a homogeneous aerosol to a fraction of the initial number, N_0, may be computed using the equation,

$$\frac{1}{N} - \frac{1}{N_0} = Kt,$$

where K in air at 68°F (20°C) has a value of 3×10^{-10} cm^3 per second (Sinclair, 1950a). The coagulation constant, K, is a function of Boltzman's constant, the viscosity of the dispersion medium and the absolute temperature.

Coagulation of an aerosol cannot be prevented by a stabilizer as in the case of liquids or solids dispersed in a liquid medium (Rodebush, 1950). In liquid aerosols the particles on collision will coalesce to form larger droplets. These large drops will tend to increase in size at the expense of the small droplets, which have a somewhat higher vapour pressure than that of the parent liquid. For this reason the droplet size in a fog is usually larger than 5 μ. In the case where the aerosol is derived from a hygroscopic liquid, such as H_2SO_4, an equilibrium is set up between the vapour pressure of the liquid droplet and the partial pressure of the water vapour in the air. The particle size of such an aerosol will depend on the humidity of the atmosphere. The particles will be smaller and more concentrated under lower humidity conditions. Solid particles may form the nuclei upon which a vapour may condense. Under this condition the solid particle itself may dissolve. Salts from sea-water sprays could form aerosols of this nature.

Settling and Dispersion

In a controlled aerosol two forms of settling may be noted: tranquil settling, where no convection currents are present, and stirred settling, where the aerosol is kept mixed at all times. Stokes' law may be used to calculate the velocity of fall, V, in centimetres per second, for various spherical particles. The applicable equations are given in Table 9 in relation to the particle-size range. This law is valid for the larger spherical particles from less than 100 μ to about 1 μ but when the size of the particles becomes comparable to the mean free path of the air molecules some correction must be applied. Under mixing conditions the rate of settling as well as the number concentration decreases exponentially with time (Sinclair, 1950a).

In non-homogeneous aerosols, such as those encountered in the outdoor atmosphere, the settling processes are much more difficult to analyse owing to prevailing meteorological conditions and to the numerous sizes and shapes of the particles that may exist. When a cloud of dust reaches the ground the particles will settle out at a rate depending on their mass and size. The concentration found at ground level depends on the total mass rate of emission and not on the concentration in the stack gas. The rate of deposition and concentration at ground level can be changed by altering the stack height. Some estimate of the rate of deposition of an aerosol is obtained by a dustfall measurement, but this measure does not evaluate the pollution that contributes to low visibility (Johnstone, 1952).

Light Scattering

In the late 19th century Lord Rayleigh (1899) devised a theory to explain the blue colour of the sky. It was based on the light scattering of extremely small transparent particles such as air molecules. He showed that the effective scattering area of a particle was a function of the volume and refractive index of the particle, and of the wave-length of the light. From his equation, for spherical particles of radius less than 0.1 of the wave-length of the light, the total scattered energy varies directly as the sixth power of the radius, and inversely as the fourth power of the wave-length, so that blue light is scattered much more than red.

Gustave Mie in 1908 developed from Clerk Maxwell's equations the theory of scattering by a spherical particle. Since that time, numerous calculations have been made of the total energy and the angular distribution of the intensity of light scattered by both transparent and absorbing particles. The derivation of the equations is given in a compact form by Stratton (1931). In the Rayleigh theory as much light is scattered backward as forward. For particles of larger radius the forward scattering is larger than the backward. When the wave-length of light is equal to the particle radius the ratio of forward to backward scattering is about 1000 or more.

When the distance between the individual particles of an aerosol is ten or more times the particle radius, the particles scatter light independently of each other. In an aerosol where the ratio of space distance to particle radius is less than about ten, interference in the scattering of light by neighbouring particles occurs (Sinclair, 1950b).

The basic physics of light scattering by small suspended particles and its relation to visibility is discussed by Sinclair (1950b). One of the most important characteristics of small suspended particles is the lowering of visibility by scattering of light. The amount of light scattering depends more on the particle size and the refractive index of the material than on the number of particles per unit volume. For most liquid aerosols the size which is most effective for scattering of light or obscuration of visibility is in the diameter range of 0.3-0.6 μ (Sinclair & La Mer, 1949). Larger particles are less effective in scattering light but may reduce visibility by absorption of light (Katz, 1954).

The haze that may be observed over a heavily industrialized area is associated with particles with diameters near the lower range of the wavelength of visible light. Increases in the concentration of such particles lower visibility. For a very dilute aerosol with a visibility perhaps of 3 miles, the concentration would be about 0.1 mg/m^3 or 45×10^6 particles (0.5 μ in diameter) per cu. ft, assuming particles of unit density. If the particle concentration increased tenfold, the visibility would be lowered to about one-third of a mile with a mass concentration of 1 mg/m^3. In the case of fog where the droplets are much larger, 5-50 μ in diameter, the number concentration would seldom be more than 140 000 droplets per cubic foot. A fog containing 200 mg/m^3 has a visibility of about 500 feet (Houghton & Radford, 1938).

Absorbance of Filtered Aerosols

Fluctuations in atmospheric particulate pollution levels in an urban area reflect the influence of changing meteorological conditions and for proper correlation with meteorological variables should be measured continuously over short time intervals. For comparison with the rise and fall of gaseous contaminants it is also essential that particulate levels such as smoke or haze concentrations be measured on a comparable time sampling basis.

In current air pollution studies some estimate of the hourly levels of airborne particulate matter is obtained by measuring the optical absorbance of the stains produced on a constant area of clean white filter paper when a known volume of air has been filtered. The extent of darkening or soiling of the surface is measured by light transmittance or reflectance and the results are converted to an index based on the relationship between the absorbance of the stain and the linear size of air sample filtered.

Indices to indicate haze or smoke concentrations or the soiling properties of air based on a linear relationship between the above parameters have been proposed by S. R. Hall (1952) and by Hemeon, Haines & Ide (1953). The Km value defined by Hall relates the absorbance of the deposit on filter paper to the mass equivalent per cubic metre of air. The Coh unit, extensively used in North America and originally proposed by Hemeon, is defined as that quantity of solids producing an absorbance of 0.01. To express a concentration, the Coh values are reduced to multiples of 1000 linear feet of air drawn through the filter paper. By definition, Km = 3.29 Cohs/1000 linear feet.

In Great Britain, Owens (Shaw & Owens, 1925) developed a method in which the deposits on a filter are compared with standard shades of colour in shade numbers from zero to 25.0. These shades are calibrated in terms of mass concentration, one shade unit being equivalent to 0.32 milligrams of smoke per cubic metre of air.

In the Detroit-Windsor air pollution studies it was found by Katz, Sanderson & Ferguson (1958) that the relationship between stain density and air quantity was curvilinear and could be expressed by an equation of the form $L = a(D \times 100)^b$, analogous to that proposed by Watson (1948) for coal dust particles in mine air. L is the length of air sample in feet, or the volume of air sample divided by the area of the filtered deposit; D is the absorbance of the stain; a and b are constants. The value b was found experimentally to be about 1.5.

In order to compare the soiling properties of polluted air in one area with that of another, all absorbance values are reduced to a basis of 1000 linear feet, then

$$1000 = a\, D^{1.5}$$

$$D = \frac{1}{a}\,(1000)^{0.67}$$

where D refers to the absorbance of a stain when the air sample is 1000 linear feet.

Also $$a = \frac{(L)^{0.67}}{D_L}$$

where D_L is the density of a stain produced by any length of air sample L.

Substitution of the above value for a in the preceding equation gives

$$D = \frac{100 \times D_L}{(L)^{0.67}}$$

Multiplication of the absorbance by 100 yields values for easy tabulation and comparison with other data (Katz & Sanderson, 1958).

The use of the linear relation does not permit of accurate comparison of soiling potential of the air from one urban area to another even when the linear air flow rate and sampling time are constant for the two areas. This is apparent from a consideration of the nature of the curvilinear equation and two areas of high and low particulate loading.

Initially the absorbance increases fairly rapidly with air quantity as the dust particles cover more and more of the filter surface until a sufficient number of particles have been collected to obscure the entire exposed area. During this process of collection there is a tendency for the particles to distribute themselves about preferred areas of the filter, as described by Dawes (1954). After the deposit has been built up in this way the absorbance will tend to increase much less rapidly with continued air sampling and dust deposition. For this reason it is a matter of general practice to institute a sampling cycle in field work such that the light transmission of the stain does not fall below 50 % of the value for the unexposed filter, or an optical absorbance of 0.301.

From the above, then, a plot of absorbance of a stain versus air quantity gives a curve, the slope of which is dependent on the particulate loading, among other factors. In the case of heavy particulate loading the absorbance rises rapidly, at first, with air quantity. As the sampling continues this rise per unit of air quantity sampled becomes increasingly smaller. With low particulate loading the rise in absorbance is at a slower rate but follows the same pattern as described above. If the soiling potentials of these two samples of air are compared by converting the respective absorbancies to some unit air quantity, assuming a linear relation, the value obtained for the heavy particulate loading will be more depressed than that for the low loading. The curvilinear relationship (the last equation) corrects for this discrepancy and follows more closely the path of the absorption versus air quantity curve for a particular air.

Condensation Nuclei and Ions

In the absence of foreign particles a highly supersaturated vapour is required before condensation will occur. A supersaturation of about 4.2 times is required for water vapour (La Mer, 1952). Very little supersaturation is required in the presence of particles that can act as nuclei for the condensation.

Nuclei are usually of natural origin, such as particles released in volcanic eruptions, in ocean sprays and in the combustion products of forest fires. Other sources may include the emissions from industrial operations, domestic heating, and transportation equipment.

It is estimated that the quantity of nuclei produced, when an acre (0.4 hectare) of average grassland is destroyed by fire, would be about 2×10^{22} (Neuberger, 1948). Coste & Wright (1935) found that when a commercial coal gas flame burned for 15 seconds the number of nuclei in a chamber increased from 109 000 to 860 000 per cubic centimetre. Amelung & Landsberg (1934) found over 500 000 nuclei per cubic centimetre in a ventilated kitchen where a large gas range was operating. The outside air in the immediate vicinity contained about 25 000 nuclei per cubic centimetre.

The size range of most nuclei is about 0.001-0.1 μ. Their weight has been estimated at 10^{-14}-10^{-18} grams or an equivalent aggregate of about 10^6 molecules (Neuberger, 1948). The size of water vapour nuclei in a sufficiently supersaturated vapour for condensation and in the absence of foreign matter to cause self-nucleation would be equivalent to an aggregate of 80 molecules of water (La Mer, 1952).

In the ordinary atmosphere the number of nuclei exceeds the number of dust particles by a factor of several thousand (Dorffel, Lattan & Rotschke, 1937). This is due to the fact that the larger dust particles are subject to considerable sedimentation and also that more nuclei are produced than dust particles in many condensation processes.

Molecules or molecular complexes of the atmospheric constituents that have lost or gained a negative charge, an electron, are termed ions. The removal of an electron from a particle requires some external energy. Such energy may come from radioactive substances such as radium and thorium, radioactive gases such as radon and thoron, cosmic rays, electric discharges, friction electricity produced by blowing sand or drifting snow, splashing water from rain showers or waterfalls, short-wave ultraviolet rays, X-rays and combustion processes (Gish, 1949; Gunn, 1951; Hagenguth, 1951). It is interesting to note that cosmic rays are practically the sole sources of ions in air over the oceans.

The number of pairs of ions produced per cubic centimetre of air near the ground in one second is about 20. The emanation of radioactive gases from the ground varies with soil porosity, soil humidity, soil temperature, ground coverage, air pressure, solar radiation, etc. The concentration of ions in the air at ground level depends not only on the rate of emanation from the ground but also on the rate of dissipation, which in turn depends on the thermal stratification and the movement of the air.

An electron may be lost or gained by a gaseous molecule or a small cluster of molecules, resulting in electrically charged condensation nuclei, and in this way a small ion may become a large ion. Large haze or dust particles and fog or cloud droplets electrostatically charged represent the ultralarge ions, and commonly carry several elementary charges either positive or negative (Gunn, 1951).

The ratio of ion velocity (centimetres per second) to the electric field intensity (volts per centimetre) is known as ion mobility and is measured in square centimetres per second volt. Ion mobility is inversely proportional to the density of the air (Gish, 1949); however, this factor is negligible in comparison to the effects on mobility produced by foreign matter in the air, particularly water vapour. Increasing concentrations of foreign material and water vapour usually diminish ion mobility, owing to the association of these materials with ions and the resultant increase in their mass.

Great variations with time and place are noted in the concentration of small ions, large ions and condensation nuclei, from which the large ions are derived. The concentration of positive ions near the ground is about 10 %-20 % higher than that of negative ions, owing partly to the attraction of positive ions and repulsion of negative ions by the negatively charged earth's surface.

The concentration of small ions over land and ocean is about the same even though the production of small ions is 15-20 ion pairs per cubic centimetre over land compared to 2 ion pairs per cubic centimetre over the ocean. This is due to the higher concentration of condensation nuclei over land. H. Landsberg (1938) summarized many condensation nuclei measurements made at different localities; and these are shown in Table 10. The much larger concentrations of condensation nuclei and of large ions over land than over the ocean reflect the efficacy of man-made and natural condensation processes as nuclei sources (Neuberger, 1948).

TABLE 10

CONDENSATION-NUCLEI CONCENTRATIONS AT VARIOUS TYPES OF LOCALITIES

Locality	Average per mm^3	Absolute maximum per mm^3
Oceans	1	40
Islands	10	109
Mountains:		
<1 km	6	155
1-2 km	2	37
>2 km	1	27
Country:		
Seashore	10	150
Inland	10	336
Towns	34	400
Cities	147	4 000

Source: Landsberg (1938).

Particulate Matter, Deposited Matter or Dustfall

The lower atmosphere varies markedly in its content of gaseous and suspended impurities from place to place. It is cleanest over the ocean. If this standard is taken as unity, then the average pollution of rural air would be 10 times greater, pollution over small towns would be about 35 times as much, and over cities pollution would be 150 times greater than that of ocean air (Landsberg, 1951). In fact, under unfavourable meteorological conditions and with large industrial sources of pollution, the city levels of pollution may be several thousand times greater than the values found over the oceans. The larger particles of smoke and dust eventually settle out and can be measured in dustfall containers. The contributed wind-blown dust of natural origin, such as sand and soil particles, is not likely to exceed the smoke and dust from chimneys in city areas by more than about 10 %, except during large dust storms or when smoke is brought in from distant forest fires.

The dustfall distribution in cities is a valuable indication of the amount of fly ash and dust deposited from stack emissions. The results are usually expressed in tons per square mile per month and serve to indicate sources and areas of high and low contamination. Most of the particles collected in this manner are larger than 20-40 μ in size. It is natural to expect, therefore, that such data will reflect the scale of industrial and domestic activity based on combustion of solid fuels and processes involving coarse dust emission. A comparison over a period of time at selected stations will indicate the success or failure of pollution abatement efforts directed towards control of excessive emission. Values of 50-100 tons per square mile[1] per month or more are found usually in the most contaminated districts of the larger, more heavily polluted cities which burn large quantities of coal.

The solid impurities collected in dustfall cans or jars consist of water-soluble and insoluble components, tarry and organic combustible material and ash. The soluble fraction in heavily polluted areas is high in sulfate and the solution is relatively acidic or low in pH value. The distribution of these components of deposited matter from samples collected in Greater Windsor and some small towns of Ontario is shown in Table 11 (International Joint Commission, Technical Advisory Board on Air Pollution, 1959). Each dustfall station is indicative roughly of conditions over an area of about 1/4 mile radius.

There is a distinct seasonal variation in deposition of total solids, from lower values in summer months to higher rates in winter months, as influenced by increased fuel consumption for heating purposes. On a percentage basis the insoluble solids constitute over 80 % of the total material collected during the summer months. This figure decreases to about 67 % in the winter period. There are corresponding changes in the percentages of the water-soluble components, as shown in Table 12. The pH of the water soluble fraction is distinctly more acidic in the winter months than in the summer period.

It is useful to compare the Windsor dustfall values with data obtained in other cities. However, this cannot be done directly because the results

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[1] 1 sq. mile = 2.6 km^2.

TABLE 11

DEPOSITED MATTER (DUSTFALL) IN THE GREATER WINDSOR AREA AND SOME TOWNS OF ONTARIO : YEARLY MEAN VALUES IN SHORT TONS PER SQUARE MILE PER MONTH *

Test period	Total solids	Insoluble (water)	Soluble (water)	Tar (CS_2)	Other combustible matter	Ash	pH
Area of relatively heavy pollution							
1951 (Aug.-Nov.)	92.2	69.2	21.1	1.48	21.0	45.9	5.4
	32.3	*24.2*	*7.4*	*0.52*	*7.4*	*16.1*	
1952 (May-Dec.)	74.3	58.9	15.4	1.39	15.0	42.1	6.5
	26.0	*20.6*	*5.4*	*0.49*	*5.3*	*14.7*	
1953	88.4	66.3	21.6	0.68	20.7	41.7	5.8
	31.0	*23.2*	*7.6*	*0.24*	*7.3*	*14.6*	
1954	96.9	64.2	32.8	0.27	22.9	36.8	5.2
	33.9	*22.5*	*11.5*	*0.09*	*8.0*	*12.9*	
1955	83.1	58.3	24.5	—	22.0	37.4	5.7
	29.1	*20.4*	*8.6*	—	*7.7*	*13.1*	
Area of relatively moderate pollution							
1951 (Aug.-Nov.)	50.9	40.9	11.3	0.59	12.1	26.2	6.4
	17.8	*14.3*	*4.0*	*0.21*	*4.2*	*9.2*	
1952 (May-Dec.)	49.0	39.6	9.3	1.37	9.5	29.0	7.3
	17.2	*13.9*	*3.3*	*0.48*	*3.3*	*10.2*	
1953	59.1	46.8	12.1	0.69	11.5	31.3	6.2
	20.7	*16.4*	*4.2*	*0.24*	*4.0*	*11.0*	
1954	65.3	44.7	20.6	0.37	11.8	33.2	5.5
	22.9	*15.7*	*7.2*	*0.13*	*4.1*	*11.6*	
1955	47.4	33.2	13.7	—	9.0	25.0	6.0
	16.6	*11.6*	*4.8*	—	*3.2*	*8.8*	
Area of relatively low pollution							
1951 (Aug.-Nov.)	35.9	26.9	8.8	0.61	11.0	16.1	6.8
	12.6	*9.4*	*3.1*	*0.21*	*3.9*	*5.6*	
1952 (May-Dec.)	31.1	26.6	6.1	0.97	5.4	19.0	7.1
	10.9	*9.3*	*2.1*	*0.34*	*1.9*	*6.7*	
1953	38.3	28.2	10.3	0.48	7.0	19.6	6.4
	13.4	*9.9*	*3.6*	*0.17*	*2.5*	*6.9*	
1954	42.4	28.4	14.2	0.11	7.6	20.8	5.8
	14.9	*9.9*	*5.0*	*0.04*	*2.7*	*7.3*	
1955	35.6	26.4	9.3	—	8.2	18.3	6.2
	12.5	*9.2*	*3.3*	—	*2.9*	*6.4*	

TABLE 11 *(continued)*

DEPOSITED MATTER (DUSTFALL) IN THE GREATER WINDSOR AREA AND SOME TOWNS OF ONTARIO: YEARLY MEAN VALUES IN SHORT TONS PER SQUARE MILE PER MONTH *

Test period	Total solids	Insoluble (water)	Soluble (water)	Tar (CS_2)	Other combustible matter	Ash	pH
Mean values for Harrow, Kingsville and Simcoe, Ont.							
1953 (Aug.-Dec.)	32.4 *11.3*	22.4 *7.8*	10.0 *3.5*	0.17 *0.06*	5.5 *1.9*	16.8 *5.9*	6.6
1954	27.6 *9.7*	16.8 *5.9*	10.9 *3.8*	0.14 *0.05*	4.2 *1.5*	12.4 *4.3*	6.1
1955	24.1 *8.4*	14.9 *5.2*	8.6 *3.0*	0.25 *0.09*	4.0 *1.4*	10.7 *3.7*	6.2

* The figures in italic type represent metric tons per square kilometre per month.

TABLE 12

SEASONAL VARIATION OF TOTAL SOLIDS, INSOLUBLE AND SOLUBLE COMPONENTS IN GREATER WINDSOR DUSTFALL, OVER PERIOD MAY 1952-MARCH 1956

Season	Total solids (tons/sq. mile/month) *a*	Water insoluble solids (%)	Water soluble solids (%)	pH Water solution
Area of heavy pollution				
Summer	58.2 *20.4*	81.5	17.6	6.1
Winter	101.5 *35.6*	67.1	33.0	5.5
Area of moderate pollution				
Summer	40.3 *14.1*	85.3	14.5	6.5
Winter	67.3 *23.6*	68.8	31.1	5.9
Area of low pollution				
Summer	28.2 *9.9*	85.5	14.3	6.6
Winter	42.2 *14.8*	66.6	33.4	6.0

a The figures in italic type represent metric tons per square kilometre per month.

are influenced greatly by the shape and dimensions of the collector used, even when there is no undue influence from other variables involved in the location of sampling stations with respect to nearby buildings and stacks. Some solution to this problem has been found in the studies carried out by Fisher (1957) at the University of Toronto. He has compared dustfall collections at identical sites with various types of container, including the Detroit-Windsor type, the British standard, the type used in the city of Toronto, and a simple stainless steel can recommended by the US Air Pollution Control Association. At Windsor, a comparative study has been made of the dustfall values obtained with the type of collector normally used in the Detroit-Windsor study area and a polyethylene jar conforming to American Pollution Control Association standards. From all this work a series of factors have been obtained to correct the dustfall data reported for various cities so as to make a more valid comparison with the Windsor-Detroit dustfall. This comparison is presented in Table 13. No allowance has been made for the fact that most cities report only insoluble solids as dustfall or sootfall. The known exceptions to this in the table are the results reported for Windsor, Toronto and the cities in the United Kingdom, where both soluble and insoluble fractions are included.

Smoke and Suspended Matter

Smoke and suspended particulate matter consist of small particles which are generally less than 1 μ in size. The particles have a low settling velocity, are readily transported by wind currents, and in the smaller sizes behave almost like gas molecules. They are generally collected by filtration, impaction, thermal or electrostatic precipitation methods in air pollution community surveys. The furnaces of the iron and steel industry, the non-ferrous metallurgical industry, the manufacture of sulfuric acid, and foundry and smelting operations are some of the sources of suspended particulate matter, in addition to smoke from the combustion of fuels. True smokes and many metal oxide fumes, such as those of lead, zinc, arsenic, cadmium, and beryllium, contain particles which are mainly in the submicron range.

The concentration of suspended particulate matter is a valuable index of pollution of the air arising from human activities as well as natural sources. Variations in concentration may be closely related to meteorological conditions and human habits. Observations in many cities indicate a daily cycle with a diurnal rhythm, one maximum occurring in the morning at about 8-9 o'clock and the other in the late afternoon or evening. This diurnal variation reflects the influence of solar heating and cooling, turbulence and other factors (Davidson, 1942; Katz, 1952c; Shaw & Owens, 1925). It has been observed in such widely separated areas as Glasgow, Leicester and other

TABLE 13

DEPOSITED MATTER IN SHORT TONS PER SQUARE MILE PER MONTH FOR A NUMBER OF CITIES *

City	Year	Dustfall reported	Factor	Dustfall corrected [a]
North America				
Detroit, Area 1	1956	73.4 *25.7*	1.00	73.4 *25.7*
Area 2		78.4 *27.5*		78.4 *27.5*
Area 3		49.2 *17.2*		49.2 *17.2*
Windsor, Industrial	1955	83.1 *29.1*	1.00	83.1 *29.1*
Residential-commercial		47.4 *16.6*		47.4 *16.6*
Residential-semi-rural		35.6 *12.5*		35.6 *12.5*
Toronto, Industrial	1956	46.1 *16.1*	1.85	85.3 *29.8*
Industrial-residential		23.6 *8.3*		43.7 *15.4*
Residential-semi-rural		11.6 *4.1*		21.5 *7.6*
New York	1956	69.8 *24.4*	1.23	85.5 *30.0*
	1955	63.8 *22.3*		78.2 *27.4*
Chicago	1947	61.2 *21.4*	1.23	75.0 *26.3*
Pittsburgh	1951	45.7 *16.0*	1.23	56.0 *19.7*
Cincinnati	1946	34.0 *11.9*	1.23	41.6 *14.6*
Los Angeles	1948	33.3 *11.7*	1.23	40.8 *14.4*
Rochester	1942	26.4 *9.2*	1.23	32.3 *11.3*
Great Britain				
Birmingham	1954	50.4 *17.7*	1.85	93.2 *32.7*
Bristol	1954	29.7 *10.4*	1.85	54.9 *19.2*
Glasgow (East)	1954	39.4 *13.8*	1.85	72.9 *25.5*

TABLE 13 *(continued)*

DEPOSITED MATTER IN SHORT TONS PER SQUARE MILE PER MONTH FOR A NUMBER OF CITIES *

City	Year	Dustfall reported	Factor	Dustfall corrected [a]
Leeds (Park Square)	1954	49.4 *17.3*	1.85	111.4 *32.0*
London (Westminster)	1954	60.2 *21.1*	1.85	97.1 *39.0*
Manchester (Phillips Park)	1954	52.5 *18.4*	1.85	 *34.0*

Sources: Great Britain, Department of Scientific and Industrial Research (1955); A.F. Fisher, personal communication, 1957; McCabe et al. (1949); New York, New Jersey & Connecticut, Interstate Sanitation Commission (1958).

* The figures in italic type represent metric tons per square kilometre per month.

[a] Dustfall values for other cities have been corrected to correspond roughly to the data obtained with the type of collector used in Detroit-Windsor.

cities in Great Britain, and New York, western Pennsylvania, and the Detroit-Windsor region.

The particulate loading of the atmosphere for a number of cities is indicated in Table 14. During periods of smog, when pollutants accumulate under the inversion layer, the concentration of smoke and suspended particulate matter may rise considerably above the daily mean. In the 1952 smog episode in London, which caused the number of deaths to rise by more than 4000 above the average for the time of year, the daily mean

TABLE 14

CONCENTRATION OF SUSPENDED PARTICULATE MATTER IN MILLIGRAMS PER CUBIC METRE IN THE ATMOSPHERE OF A NUMBER OF COMMUNITIES

Community	Minimum	Maximum	Mean
Baltimore, Centre	0.21	1.72	0.87
Industrial area	0.05	1.46	0.38
Cincinnati, Industrial area	0.01	1.98	0.42
Residential area	0.01	1.30	0.28
Donora	0.00	2.50	0.74
Los Angeles, Detroit, San Francisco, Washington	0.25	0.47	0.51
Windsor, Areas of high pollution [a]	0.036	0.47	0.21
Areas of moderate pollution	0.001	0.56	0.15
Areas of low pollution	0.003	0.21	0.08

Source: Cholak (1952); Katz (1952b). (By permission from the Stanford Research Institute.)

[a] October-December 1951 (Katz, 1954).

levels of smoke or fine particulate matter in the air increased to a maximum of 4.46 mg/m^3 during the period 3-10 December, or about 14 times the daily mean recorded on 11 December (National Smoke Abatement Society, 1953).

More recently a considerable body of information on the particulate loading and composition of the atmosphere has been made available through the operation of the National Sampling Network of the US Public Health Service (Chambers, Foter & Cholak, 1955). For example, the air samples from five cities having populations in excess of 2 million within their corporate limits exhibit relatively high levels of particulate matter. The average total particulate loading for this class of city was reported to be 264 μ/m^3. On the other hand, the corresponding loading for a group of cities of intermediate population range, between 500 000 and 2 million, was about half that of the large-city group—namely, 137 μ/m^3. The results from other urban areas served to confirm the qualitative observation that the concentration of suspended particulates in the air over a community is, very broadly, related to the number of persons contributing to the pollution.

Chemical Composition of Particulate Pollution

Aerosol contaminants in city air are extremely complex in chemical composition. More than 20 metallic elements have been found by chemical and spectrographic analysis of the inorganic fraction, in addition to carbon or soot and tarry organic material. The most abundant metallic elements are silicon, calcium, sodium, aluminium, and iron. Relatively high quantities of magnesium, lead, copper, zinc, and manganese may also be found. The concentrations of these elements will depend upon the nature of the principal industries and the effectiveness of measures taken to control particulate emissions (Cholak, 1952). The distribution of lead in the air of cities has been correlated with density of vehicular traffic, because lead compounds are widely used as anti-knock substances in gasoline (Cholak, 1952; E. L. Hall, 1952).

The maximal concentrations of various components of particulates encountered in the US national air pollution survey are shown in Table 15. These values represent the extreme 24-hour integrated dosages prevalent under the most adverse circumstances encountered in an 18-month survey and can occur only very rarely. Average or median concentrations of components for large cities of over 2 million population are given in Table 16, values for cities of intermediate range in Table 17, and corresponding data for non-urban areas in Table 18 (Chambers, Foter & Cholak, 1955).

There are evident qualitative differences in the fractional analyses that may be indicative of dominant sources of pollution in each area. For

TABLE 15

MAXIMAL URBAN AIR LOADINGS (US NATIONAL AIR SAMPLING NETWORK)

Particulate matter	$\mu g/m^3$	Particulate matter	$\mu g/m^3$
Total	3 112.0	Mg	15.5
Acetone-soluble	234.0	Na	23.7
Proteins	110.6	K	8.7
Mn	5.70	Sr	1.55
Pb	26.6	Be	0.0030
Sn	0.84	As	1.41
Fe	53.6	F^-	1.64
Cu	4.68	Cl^-	7.6
Ag	0.0746	SO_4^{--}	87.8
Ti	2.57	NO_3^-	40.4
V	0.640	Polycyclic hydrocarbons	29.6
Cr	3.5		
Zn	0.78		

Source: Chambers, Foter & Cholak (1955). (By permission from the Stanford Research Institute.)

TABLE 16

PARTICULATE ANALYSES (MICROGRAMS PER CUBIC METRE) FROM CITIES HAVING POPULATIONS OVER 2 000 000 (US NATIONAL AIR SAMPLING NETWORK)

Particulate matter	Los Angeles	Detroit	Philadelphia	Chicago	New York
Total load	265	344	188	280	244
Acetone-soluble	57.3	50.9	30.0	45.8	37.7
Mn	0.15	0.74	0.54	0.11	0.07
Pb	5.2	2.9	9.5	2.8	2.8
Sn	0.02	0.06	0.16	0.03	0.08
Fe	4.7	8.3	6.5	7.9	5.2
V	0.002	0.025	0.174	0.002	0.322
Cu	0.13	0.57	1.34	0.11	0.30
Be	0.0001	0.0004	0.0005	0.0002	0.0003
Ti	0.30	0.28	0.15	0.18	0.41
As	0.02	0.04	0.16	0.04	0.05
F^-	0.38	0.04	0.24	0.03	0.21
SO_4^{--}	14.4	7.5	15.6	9.4	14.8
NO_3^-	14.4	1.2	1.6	1.7	0.8

Source: Chambers, Foter & Cholak (1955). (By permission from the Stanford Research Institute.)

example, Los Angeles shows almost twice as much acetone-soluble matter as Philadelphia and exceeds all other cities of the network in this respect. Another striking feature of the Los Angeles samples is the relatively high value for nitrate ion. Lead is another toxic constituent which is relatively

TABLE 17

PARTICULATE ANALYSES (MICROGRAMS PER CUBIC METRE) FROM CITIES HAVING POPULATIONS BETWEEN 500 000 AND 2 000 000 (US NATIONAL AIR SAMPLING NETWORK)

Particulate matter	Cincinnati	Kansas City	Portland	Atlanta	Houston	San Francisco	Minneapolis
Total load	176	146	143	137	129	104	120
Acetone-soluble	31.4	18.4	32.1	24.2	18.5	19.4	15.8
Mn	0.24	0.08	0.23	0.12	0.23	0.11	0.08
Pb	1.6	1.0	1.2	1.8	1.0	2.4	0.5
Sn	0.03	0.03	0.01	0.03	0.02	0.02	0.01
Fe	4.5	4.1	5.1	3.3	4.0	2.4	4.4
V	0.09	0.002	0.009	0.024	0.001	0.002	0.002
Cu	0.18	0.04	0.05	0.01	0.02	0.07	0.60
Be	0.0002	0.0003	0.0003	0.0002	0.0002	0.0001	0.0002
Ti	0.06	0.21	0.24	0.12	0.29	0.04	0.11
As	0.02	0.02	0.02	<0.01	0.01	0.01	0.01
F^-	0.21	0.01	Nil	0.05	Nil	0.37	0.06
SO^{--}_4	5.6	1.5	0.8	1.0	2.4	1.8	0.8
NO^-_3	1.0	0.6	0.2	0.8	1.0	3.4	1.3

Source: Chambers, Foter & Cholak (1955). (By permission from the Stanford Research Institute.)

prominent in both Philadelphia and Los Angeles. The highest values for sulfate ion were found in Philadelphia, New York and Los Angeles.

Spectrographic analyses of 23 samples of particulates collected in the

TABLE 18

PARTICULATE ANALYSES (MICROGRAMS PER CUBIC METRE) FROM NON-URBAN AREAS (US NATIONAL AIR SAMPLING NETWORK)

Particulate matter	Boonsboro	Salt Lake City	Atlanta	Cincinnati	Portland
Total load	68	55	71	45	86
Acetone-soluble	8.7	6.2	9.3	9.0	12.6
Mn	0.00	0.04	0.11	0.07	0.04
Pb	0.1	0.1	0.9	0.4	0.3
Sn	<0.01	<0.01	<0.01	0.01	<0.01
Fe	3.7	4.1	2.7	2.4	3.6
V	0.003	Nil	0.004	0.001	0.002
Cu	Nil	0.28	<0.01	0.19	<0.01
Be	0.0001	<0.0001	0.0002	0.0001	0.0001
Ti	0.26	Nil	0.13	0.01	Nil
As	0.01	0.03	0.01	<0.01	0.04
F^-	—	—	Nil	0.26	—
SO^{--}_4	0.3	<0.01	0.5	1.9	0.4
NO^-_3	—	—	—	0.7	—

Source: Chambers, Foter & Cholak (1955). (By permission from the Stanford Research Institute.)

Windsor area yielded the following mean values for elemental components, in micrograms per cubic metre of air: silicon 6.2; calcium 3.3; aluminium 2.8; iron 2.5; magnesium 0.8; and lead 0.6 (Katz, 1952c). Other elements such as Mn, Cu, and Zn were present in concentrations of less than 0.3 μ/m^3. Heavy metals such as Ti, Sn, Mo, Ba, Ni, V, and Cr showed mean concentrations in the range of less than 0.1; Cd and Be were present only in trace quantities with mean values of less than 0.01. These results are in substantial agreement with those reported for the US Air Sampling Network for cities in the population class of Windsor.

The application of X-ray diffraction technique to air pollution problems has proved itself useful in the study of air-borne contaminants in the Windsor area. The unique feature of the method is that, unlike chemical, polarographic and spectrographic methods, it reveals the state of chemical combination of the crystalline constituents of particulates. A limitation of this technique is that it gives no information about amorphous materials. However, the various means used to separate specimens from a sample of dust often afford indications of relative abundance of constituents both crystalline and amorphous. Constituents identified in air-borne particulates collected in the Windsor area include calcium carbonate, silica, aluminium metal particles, gypsum, ammonium chloride, magnetite, hematite, nickel fluoride, cadmium carbonate, graphite, carbon, brass, and solder particles (Shore & Katz, 1954).

In nearly all of the above samples, the strongest diffracting constituents were α-quartz and calcium carbonate. The X-ray diffraction studies showed fair agreement with the spectrographic analyses.

Composition of Organic Fraction

The organic constituents of atmospheric pollution have been receiving increasing attention in recent years because of their importance from a health standpoint, especially the aromatic hydrocarbons that are potent carcinogens. In the Los Angeles area, the hydrocarbons represent one of the largest single groups of pollutants discharged to the atmosphere, and a considerable number of saturated and unsaturated hydrocarbons in the C_2 to C_{10} range have been individually identified by the mass spectrometer (Shepherd et al., 1951). Condensed polycyclic aromatic compounds in the organic fraction of the urban atmosphere, in particular the benzopyrenes, have been associated with the increase in the incidence of lung cancer (Kennaway & Kennaway, 1947).

Usually, the organic fraction of suspended particulates, collected on filters, is separated by solvent extraction. For protein analysis, an 8-hour extraction with acetone removes most of the non-protein organic matter.

However, for general organic chemical separation, a more useful product is provided by the extraction of samples with benzene. Purified cyclohexane is a preferred elution agent for the separation and identification of polynuclear hydrocarbons by chromatographic adsorption technique.

The variation in monthly average levels of the benzene-soluble organic matter in particulate samples from the atmosphere over a number of cities is illustrated by the data in Table 19. With the exception of Windsor, the smallest city listed in this table, the results were abstracted from air pollution measurements of the US National Air Sampling Network (US Department of Health, Education, and Welfare, Public Health Service, 1958). Some cities show a distinct seasonal trend in the organic matter content of the air, with lowest values in the summer months and maximum levels in the winter season. In other cities this trend is obscured.

TABLE 19

MONTHLY AVERAGE BENZENE-SOLUBLE ORGANIC MATTER IN VARIOUS URBAN ATMOSPHERES (MICROGRAMS PER CUBIC METRE) $\times 10^{-1}$

Months	Location and years					
	Windsor 1955-56	Detroit 1956-57	Philadelphia 1955-57	New York 1955-57	Kansas City 1955-57	St Louis 1955-57
January	78	86	174	124	206	239
February	73	141	185	151	123	312
March	49	92	161	122	90	135
April	62	73	177	118	132	94
May	45	69	145	91	74	112
June	52	117	133	82	104	109
July	40	82	106	55	60	126
August	37	76	111	79	99	127
September	40	86	125	83	93	177
October	—	210	137	142	121	254
November	—	84	180	125	108	172
December	54	113	220	90	159	264

The factor of age of samples of particulate material is of some significance in relation to products isolated subsequently. This factor is under investigation and there is evidence that low boiling-point compounds are retained by the particulate matter during transport and storage because of the strong adsorptive properties of the material on the filter. There is little or no evidence from the results of the US Network programme to indicate that any important chemical changes occur during transport or storage of samples. Soluble organic materials from particulate matter collected on glass fibre filters at sampling stations in 20 cities of the Network have been separated by Tabor, Hauser & Burttschell (1956) into the following fractions: water-soluble, water-ether-insoluble, basic, weak acid or phenolic, strong acid and neutral. The neutral group was further subdivided by chromatography into aliphatic and aromatic hydrocarbons and oxygenated derivatives.

Polynuclear Aromatic Hydrocarbons and Carcinogens

The presence of carcinogens in urban air has been reported in Great Britain, the USA, Canada and elsewhere. Kotin et al. (1954) showed that the Los Angeles air contained a number of carcinogens, including 3:4-benzpyrene. In Great Britain, Clemo & Miller (1955) demonstrated the presence of substances having marked carcinogenic action in city smoke, and Waller (1952) identified benzpyrene in the air over a number of cities. Kotin et al. (1954) and Kotin, Falk & Thomas (1954, 1955) have identified pyrene, benzpyrenes, and other polycyclic aromatic hydrocarbons in the particulates from the exhaust of both gasoline and Diesel engines. More recently, Falk & Kotin (1956) have demonstrated the production of cancerous tumours in mice by skin exposure of acetone extracts of aliphatic compounds consisting of branches and cyclic hydrocarbons and their epoxide states. The results of an examination of the Salford, Lancashire, atmosphere by Cooper (1954) revealed the following concentrations of polycyclic hydrocarbons in micrograms per 100 cubic metres of air: pyrene, 18.0-35.0; 3:4-benzpyrene, 19.7-29.0; 1:12-benzperylene, 16.1-35.0; and fluoranthene, 20.0.

Similar investigations of the Windsor atmosphere by Shore & Katz (1956) revealed the presence by specific identification of pyrene, fluoranthene, benz(a)anthracene, chrysene and benz(e)pyrene. The presence of benz(a)pyrene was also indicated, as well as higher members of this aromatic series.

Infra-red absorption data on carbon tetrachloride extracts of Windsor particulates showed strong absorption bands for the presence of aliphatic and aromatic C-H bonds, aldehyde, ketone or ester and carboxylic acid groups. The spectrum of the extract was similar, qualitatively, to those obtained by others for air samples from Detroit, Philadelphia and Houston (Chambers, Tabor & Foter, 1955).

Gases and Vapours

Sulfur dioxide

This gas is one of the principal contaminants and has been studied extensively in many urban and industrial areas. The measurement of concentration levels and study of dispersion of smoke and sulfur dioxide have been conducted systematically for many years in a country-wide network of sampling stations in Great Britain, under the guidance of the Department of Scientific and Industrial Research. These studies constitute, probably, the most comprehensive information available on the nature of the association between air pollution and fog to produce smog. A great number of local or municipal authorities participate in this work; for example, between 1944 and 1954 the number of co-operating bodies rose from 77 to 222. One of the most important recent developments in the application of the results has been the study of the distribution of pollution in relation to the incidence of respiratory diseases, including bronchitis and lung cancer.

Sulfur dioxide is readily absorbed by vegetation, soil and water surfaces, and accelerates the corrosion and deterioration of wire, metals, paper and textiles, and building materials. Many species of green plants are particularly susceptible to injury by this gas, particularly at concentrations above about 0.3 p.p.m., if exposure is prolonged for more than a few hours under optimum light, temperature, relative humidity, soil moisture and other growth conditions (Katz, 1949).

Relatively high average and maximum concentrations of sulfur dioxide have been reported in many of the larger cities of Great Britain, the USA and elsewhere. The annual consumption of coal in such cities may amount to many millions of tons, in addition to liquid and gaseous fuels. The sulfur content of the fuel varies from less than 1 % for good quality anthracite to over 4 % for some grades of bituminous (high-volatile) coal. In the USA, the annual consumption of coal in Pittsburgh is about 20 million tons, New York City nearly 32 million, Greater Detroit 13 million, St Louis 5.5 million, and Birmingham 11 million.

The average concentration varies with the amount and sulfur content of the fuel used for heat, electricity and power, and hot water. In certain areas there is a distinct seasonal variation, the average concentrations rising during the cold months of the heating season. Since the industrial consumption of fuel does not vary greatly throughout the year, this variation is due mainly to domestic use and demand for space heating. In recent years there has been a progressive decrease in the average atmospheric concentrations of several large cities owing to the increased use of coal with a lower sulfur content and natural gas or oil for space heating. Air pollution control measures in St Louis prohibit the use of high sulfur coal. The trend in North America towards increased consumption of natural gas, instead of coal, for space heating and industrial operations is likely to accelerate in the future and will result in a considerable reduction in sulfur dioxide pollution and smoke.

Comparative figures of mean sulfur dioxide levels in a number of cities in Great Britain are given in Table 20. A distinct seasonal trend is evident. Fairly extensive studies have been made of sulfur dioxide conditions in the Greater Detroit-Windsor area as the result of an air pollution reference to

TABLE 20

CONCENTRATIONS OF SULFUR DIOXIDE IN THE ATMOSPHERE OF A NUMBER OF CITIES IN GEAT BRITAIN

City and site	Sampling period	Average sulfur dioxide concentration (p.p.m. by volume)			
		Summer	Winter	Year	Highest daily mean
Glasgow	1949-54	0,06	0.11	0.09	0.86
Leeds	1950-54	0.11	0.22	0.16	0.87
Leicester	1949-54	0.06	0.14	0.10	1.17
London (Lambeth)	1949-54	0.08	0.19	0.14	1.34
London (Westminster)	1950-54	0.08	0.18	0.13	1.23
Salford	1949-54	0.10	0.20	0.15	0.95
Sheffield	1949-54	0.11	0.18	0.14	1.14
Wolverhampton	1949-54	0.02	0.05	0.04	0.24

Source: Great Britain, Department of Scientific and Industrial Research (1955).

TABLE 21

CONCENTRATIONS OF SULFUR DIOXIDE IN THE ATMOSPHERE OF THE DETROIT-WINDSOR AREA

Station	Sampling period	Average sulfur dioxide concentration (p.p.m. by volume)			
		Summer	Winter	Year	Maximum monthly mean
Detroit, Area 4	July 1953-April 1955	0.047	0.130	0.095	0.180
Detroit, Area 24	July 1953-April 1955	0.030	0.058	0.046	0.070
Windsor, High pollution	September 1950-55	0.051	0.047	0.044	0.116
Windsor, Low pollution	September 1950-55	0.011	0.020	0.016	0.073
Windsor, Semi-rural	September 1950-55	0.003	0.015	0.007	0.027

Source: International Joint Commission, Technical Advisory Board on Air Pollution (1959).

the International Joint Commission of the USA and Canada to investigate the pollution problem in the Detroit River area (International Joint Commission, Technical Advisory Board on Air Pollution, 1959). Some results of this study are shown in Table 21. Other data on concentrations in the atmosphere of a number of American cities are given in Table 22. Substantially higher levels of gas are encountered in cities of larger population where solid fuel consumption is relatively greater.

TABLE 22

CONCENTRATIONS OF SULFUR DIOXIDE IN THE ATMOSPHERE OF A NUMBER OF COMMUNITIES *

Community	Number of samples	Concentration (p.p.m.)	
		Range	Average or mean
Chicago (1937)			
Railroad station	32	Trace-2.54	0.67
Manufacturing area	99	0.17-3.16	0.50
Commercial area	89	Trace-3.16	0.41
Pittsburgh (1931-32)			
Industrial area	607	0.00-2.5	0.30
St Louis (1937)			
Cold months	663	0.0-1.86 [a]	0.25 [a]
Chicago (1937)			
Residential area	32	Trace-0.47	0.16
Donora (1949)	529	0.00->0.50	0.15 ($\sigma = \pm 0.13$)
St Louis (1937)			
Warm months	673	0.0-0.756 [a]	0.127 [a]
Baltimore (1950)			
Industrial area	79	0.01-0.46	0.074
Cincinnati (1947-51)			
Industrial and commercial areas	227	0.00+-0.46	0.064 ($\sigma = \pm 0.058$)
All areas	298	0.00+-0.46	0.061 ($\sigma = \pm 0.061$)
Residential area	71	0.00+-0.27	0.044 ($\sigma = \pm 0.058$)
Cleveland (1949-50)			
Industrial area	290	0.0-0.489 [a]	0.042 [a]
St Louis (1950)			
Cold months	243	0.0-0.51 [a]	0.041 [a]
Los Angeles (1949-50)		Trace-0.60	
Cincinnati (1947-51)			
Rural area	69	0.00-0.31	0.037 ($\sigma = \pm 0.056$)
Salt Lake City (1919-20)	27		0.036
Yonkers (1936-37)	continuous observations	0.0-0.75	0.032
St Louis (1937)			
Warm months	294	0.0-0.24 [a]	0.03 [a]
Charleston (1950-51)	143	0.001-0.18	0.023 ($\sigma = \pm 0.027$)
Baltimore (1950)			
Rural area	12	0.00+-0.11	0.023

Source: Cholak (1952). (By permission from the Stanford Research Institute.)

* Arranged in descending order.

[a] SO_2 values as such.

Greenburg & Jacobs (1956) have estimated recently that the annual emission of sulfur dioxide to the New York City atmosphere from combustion of fuels is in excess of 1.5 million tons. Studies of the seasonal variations show a peak in December and January, when the average concentration attains about 0.25 p.p.m. Diurnal variations in concentration were also noted, with a maximum usually occurring during the morning period from 6 to 10 a.m. A subsidiary maximum also was found to occur in the late afternoon or early evening. Such diurnal variations have also been observed in the Detroit-Windsor area. Since no solid fuel is used in the Los Angeles area, sulfur dioxide levels are comparatively low for a city of this size. Here also, the concentrations show a diurnal trend, with average values in the range of 0.05-0.10 p.p.m.

Hydrogen sulfide and organic sulfides

As a rule, these substances are not liberated in appreciable quantities by industrial operations in city communities. They cause odour nuisances when present in the air at concentrations 10-100 times smaller than the lowest concentration of sulfur dioxide detectable by smell. Mercaptans and hydrogen sulfide are evolved in the manufacture of coke, distillation of tar, petroleum and natural gas refining, manufacture of viscose rayon, and in certain chemical processes. Such effluents are either burned to sulfur dioxide before liberation to the air or else are absorbed in " purifiers " containing iron oxide. In Great Britain the iron sulfide or " spent oxide " from such purification measures is used for the production of sulfuric acid on a considerable scale. The recovery of hydrogen sulfide by the Girbotol and other processes is becoming standard practice in the petroleum industry and represents a source of elemental sulfur of increasing importance.

A dramatic incident involving deaths from air pollution by the escape of hydrogen sulfide from the Girbotol units of a natural-gas refining plant occurred at Poza Rica, Mexico, in November 1950. This severe fumigation caused the deaths of 22 persons and the hospitalization of about 300 others (McCabe & Clayton, 1952). The symptoms of the affected population and other circumstances indicated that the gas concentrations must have reached extremely high levels over a period of about one hour.

Although the maximum allowable concentration for H_2S is 20 p.p.m. by volume, its odour may be detectable in the range of 0.035-0.10 p.p.m. A limited number of observations in some cities have usually shown the existence of relatively low concentrations, less than 0.10 p.p.m. Of 49 air samples analysed in the Windsor area, concentrations of H_2S ranged from 0.0 to 0.61 p.p.m., with a mean of 0.11 p.p.m. (Katz, 1952c). The high values were recorded close to sources having a distinct odour.

Hydrogen fluoride and chloride

Fluorides may be present in the atmosphere in gaseous or solid forms as effluents from the aluminium industry, from the manufacture of phosphate fertilizer, brick plants, pottery and ferro-enamel works. Small amounts are also liberated in the combustion of coal and from fluxing agents used in making foundry iron and in miscellaneous cupola and blast furnace operations. Hydrogen fluoride and volatile fluorides are important air contaminants in extremely low concentrations, 0.001-0.10 p.p.m. by volume. The gas accumulates in the leaves of plants and can be absorbed in forage to such an extent as to cause fluorosis in animals. Gladiolus, prune, apricot, and peach, among other sensitive plants and flowers, are extremely susceptible to hydrogen fluoride in concentrations as low as 0.02-0.05 p.p.m. This has created a problem of special concern to agriculture in the vicinity of large aluminium refining and steel manufacturing operations. Fluorides, especially gaseous compounds, rapidly attack glass by etching the surface.

The concentrations which have been found in the air of a number of communities are shown in Table 23. The values are lower than those of virtually all other pollutants which may occur in the atmosphere from industrial activities. However, the high degree of toxicity of fluorine compounds renders the control of such emissions imperative for industries manufacturing aluminium metal and phosphate fertilizer.

Hydrochloric acid and chlorine caused widespread damage to vegetation and property in Great Britain in the early days of the alkali industry, when the by-products of the Leblanc soda process were allowed to escape into the

TABLE 23

CONCENTRATIONS OF FLUORIDE
IN THE ATMOSPHERE OF A NUMBER OF COMMUNITIES *

Community	Number of samples	Concentration (p.p.m.) as HF	
		Range	Average or mean
Baltimore (1950)			
Industrial area	79	0.000- 0.08	0.018
Residential area	21	0.000- 0.021	0.008
Cleveland (1949-50)			
Industrial area	256	0.000->0.063	0.014
Donora (1948-49)	40	0.000- 0.06	0.008
Donora (1949)	249	0.000->0.03	0.006
Los Angeles (1948)	20	0.000- 0.025	0.008
Cincinnati (1946-51)			
All areas	386	0.000- 0.025	0.005+
Rural area	75	0.000- 0.025	0.0047
Residential area	94	0.000- 0.025	0.006
Industrial and commercial areas	292	0.000- 0.025	0.005
Charleston (1950-51)	132	0.000- 0.03	0.003

Source: Cholak (1952). (By permission from the Stanford Research Institute.)

* Arranged in descending order.

atmosphere (Meetham, 1952). The first Alkali, etc. Works Regulation Act of Great Britain, passed in 1863, specified a 95 % recovery of such emissions, and an amendment to this Act in 1874 limited the discharge of hydrochloric acid to 0.2 grain [1] per cubic foot of flue gas, before dilution with other stack gas or air. However, recovery of such by-products is relatively easy and profitable, so that escapes of significant quantities seldom occur, except under accidental conditions. The modern alkali industry is based on the electrolysis of common salt, and by-products are usually carefully controlled. Average concentrations of chlorides reported for a number of cities, calculated as Cl^- in p.p.m., vary from about 0.016 to 0.095, as shown in Table 24.

TABLE 24

CONCENTRATIONS OF CHLORIDE IN THE ATMOSPHERE OF A NUMBER OF COMMUNITIES *

Community	Number of samples	Concentration (p.p.m.)	
		Range	Average or mean
Chicago (1912-13)			0.078 [a]
Donora (1949)	247	0.00 -0.3	0.071 ($\sigma = \pm 0.067$)
Donora (1948-49)	39	0.014-0.27	0.070
Baltimore (1950)	79	0.001-0.11	0.037 ($\sigma = \pm 0.028$)
Cincinnati (1946-51)			
All areas	227	0.00 -0.19	0.033 ($\sigma = \pm 0.033$)
Rural area	52	0.00 -0.106	0.026 ($\sigma = \pm 0.027$)
Residential area	51	0.00 -0.108	0.030 ($\sigma = \pm 0.027$)
Industrial and commercial areas	176	0.00 -0.19	0.034 ($\sigma = \pm 0.034$)
Charleston (1950-51)	152	0.00 -0.068	0.016
Salt Lake City (1919-20)		0.001-0.025 [a]	
Windsor, Ont.	48	0.00 -0.62	0.095

Source: Cholak (1952); Katz (1952b). (By permission from the Stanford Research Institute.)

* Arranged in descending order.

[a] Recalculated as Cl^-.

Oxides of nitrogen and ammonia

It is probable that oxides of nitrogen are the second most abundant atmospheric contaminant in many communities, ranking next to sulfur dioxide. These oxides are important by-products of the chemical industry in the manufacture of nitric acid, sulfuric acid by the chamber process, nitration of organic compounds, and manufacture of nylon intermediates, and are formed in varying quantities in internal combustion engines from the air supplied to the fuel. Significant amounts are liberated in the exhaust gases of trucks and passenger automobiles. As shown earlier, automobiles, trucks and buses in the Los Angeles area emit about 430 tons of nitrogen oxides to the atmosphere daily. In the combustion of natural gas by domestic gas appliances and furnaces, the products of combustion may contain from 15 to 50 p.p.m. of oxides of nitrogen. The most favourable conditions for the formation of these oxides in the combustion chamber of furnaces are high temperature followed by rapid cooling of the products of combustion. Industrial furnaces and internal-combustion engines may release up to 500 p.p.m. or more of nitrogen oxides in the waste gases (E. L. Hall, 1952).

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[1] 1 grain = 0.064 g.

Oxides of nitrogen, as such, are more dangerous to human health than to vegetation. Both nitric oxide and peroxide are readily absorbed by the blood, after inhalation into the lungs, and combine with the haemoglobin to form an addition complex similar to the action of carbon monoxide. In this respect, these oxides are more injurious than carbon monoxide in equivalent concentrations. It is standard practice in the chemical industry to absorb and recover significant quantities of oxides of nitrogen.

The concentration levels of oxides of nitrogen have been studied extensively in the Los Angeles area. Some indication of the observed values is given in Tables 25 and 26. The levels show a diurnal variation with a peak at about 8 a.m. (Hitchcock et al., 1955; Larson, Taylor & Hamming, 1955). These oxides have also been measured in other communities but the sampling has been carried out on a limited basis only and comparisons are not justified.

TABLE 25

CONCENTRATIONS OF POLLUTANTS IN THE LOS ANGELES ATMOSPHERE
(MAXIMUM VALUES AS MEASURED
OVER CENTRAL LOS ANGELES ON VARIOUS DAYS)

Concentrations, in p.p.m. by volume			Concentrations $\mu g/m^3$		
Pollutant	Day of good visibility	Day of reduced visibility	Pollutant	Day of good visibility	Day of reduced visibility
Acrolein	—	Present	Aluminium	0.003	0.008
Lower aldehydes	0.07	0.4	Calcium	0.006	0.007
Carbon monoxide	3.5	23.0	Carbon	0.035	0.132
Formaldehyde	0.04	0.09	Iron	0.003	0.010
Hydrocarbons	0.2	1.1	Lead	0.002	0.042
Oxidant	0.1	0.5	Ether-soluble aerosols	0.012	0.120
Oxides of nitrogen	0.08	0.4	Silicon	0.007	0.028
Ozone	0.06	0.3	Sulfuric acid	0.000	0.110
Sulfur dioxide	0.05	0.3			

Source: Los Angeles County Air Pollution Control District (1949, 1951).

TABLE 26

MAXIMUM CONCENTRATIONS OF AIR POLLUTANTS MEASURED IN LOS ANGELES COUNTY

Pollutant	Concentration (p.p.m.)	Location	Date
Total aldehydes	1.87	Pasadena	30/5/57
Carbon monoxide	72.00	Central L.A.	3/12/57
Total hydrocarbons	4.66	El Segundo	11/8/57
Nitrogen dioxide	1.74	Central L.A.	17/9/53
$NO + NO_2$	1.86	Dominguez	12/12/57
Total oxidant (KI)	0.69	Central L.A.	13/9/53
Ozone (rubber cracking)	0.90	Vernon	13/9/55
SO_2 (autometer)	2.49	El Segundo	27/6/57
Particulate matter	26.4 K_m units	Vernon	22/11/57

Source: Los Angeles County Air Pollution Control District (1949, 1951).

Ammonia and ammonium salts are not important air contaminants. Ammonia is an important raw material in the fertilizer and organic chemical industries and in the manufacture of nitric acid by the oxidation process. Its recovery is a matter of fundamental importance in the economical operation of such processes and in the manufacture of gas from coal.

Aldehydes

Lower aldehydes may be present in the atmosphere of some industrial communities in concentrations about as high as sulfur dioxide. Relatively high aldehyde levels have been reported in Los Angeles, and in some other cities such as Cincinnati and in the industrial-commercial section of Windsor, near the Detroit River (Cholak, 1952; Katz, 1952b; Larson et al., 1955). Formaldehyde is irritating to the eyes of some people in concentrations as low as about 0.25 p.p.m. by volume, although several p.p.m. are required to affect the majority of persons exposed (Los Angeles County Air Pollution Control District, 1949, 1951). In the Los Angeles district pronounced eye irritation is experienced by large numbers of people whenever intense smog conditions prevail. However, the aldehyde concentrations alone are not high enough to account for this effect.

Cholak (1952) has shown that aldehydes in the air vary not only with the nature of the activity of an area but also with the density of motor traffic. The incomplete oxidation of motor fuel and lubricating oils leads to the formation of aldehydes and organic acids. The products of combustion of natural gas may also contain aldehydes. It is probable that atmospheric contamination by aldehyde is due more to the exhaust and blow-by gases of motor vehicles than to the waste products of industrial and heating plant sources.

Carbon monoxide

Next to carbon dioxide, carbon monoxide is the most abundant pollutant and may show wide diurnal variations in the urban atmosphere. The concentrations in city streets vary with the density of motor traffic. Fairly extensive studies of this contaminant have been made by Cholak (1952) in Cincinnati and by the Los Angeles Air Pollution Control District (see Tables 25 and 26). In the commercial and industrial districts of Cincinnati, the concentrations have ranged from 0 to 55 p.p.m., with an average of 9.5 p.p.m. as compared to about 4.0 p.p.m. in the residential area.

In Los Angeles, carbon monoxide levels, in a 4-month continuous aerometric survey carried out in 1954 (Hitchcock et al., 1955), have been reported to reach a diurnal mean peak of 8.8 p.p.m. at 8.30 a.m., with hydrocarbons at 0.38 p.p.m. at the same time. The CO levels drop sharply to about 3.5 in the early afternoon and rise again to about 7.0 p.p.m. at 4.30 p.m. Corresponding values for aldehydes exhibited smaller variations about the maximum average of 0.20 p.p.m. attained at about noon. The maximum concentration of carbon monoxide in the centre of Los Angeles was reported to be as high as 72 p.p.m. in December 1957.

Organic vapour contaminants

Considerable success has been attained in the identification of condensable organic impurities in the atmosphere by mass spectrometric techniques, as devised by the late Dr Martin Shepherd of the US National Bureau of Standards. For this purpose the atmospheric pollutants are collected by condensation in a Shepherd trap cooled to the temperature of boiling oxygen (—183°C). Relatively large volumes of air must be sampled in this manner as the pollutants of primary concern here may be present in a total concentration of as little as 0.10 p.p.m. or less. About 50 chemical substances were identified in the mass spectrometer by Shepherd et al. (1951) in a single sample collected in Los Angeles in 1950.

Weaver & Gunther (1955) have reported, more recently, on the composition of samples taken in Los Angeles, Pasadena, Detroit, Wilmington and Washington. Condensates from all these cities showed mass spectra of the same general character with a considerable degree of regularity. There was a close resemblance between traffic gas and smog condensates in several cities, and this is evidence of a common genesis in motor fuels and motor traffic. Two evolutionary steps were evident—first, the reactions that occur in the engine cylinders and, secondly, those that occur in the air afterwards. The results have been compared with mass spectrometric records of several samples taken during some of the brilliant experiments of Dr Haagen-Smit with mixtures of hydrocarbons and oxides of nitrogen under ultraviolet irradiation. Weaver & Gunther (1955) state that the second group of reactions—those that occur in traffic gases when they enter the atmosphere—would be completely baffling, except for the work of Haagen-Smit (1952).

A significantly larger number of mass peaks were found after combustion of gasoline than before, and there was a general redistribution of masses. This consisted of a relative increase in ions of low masses, and in high masses, especially above 100. The percentage of benzene in condensate from traffic gas was about 1.7 times that in gasoline; of toluene 3 times, and of xylene 30 times. There was evidence of the formation of oxygen compounds and also a striking increase in ions characteristic of the heavier unsaturated compounds of the olefin or cycloparaffin series and the acetylene or diene series, especially the latter. In a general way the changes that occurred in Haagen-Smit's reaction chamber were similar to the changes necessary to convert the mass pattern of a typical traffic gas sample into a typical Los Angeles smog pattern. A number of ions were identified as oxides or epoxides of hydrocarbons formed through the oxidation of unsaturated hydrocarbons.

It was not possible to identify more than a small fraction of the large number of chemical compounds present in the vapour state. About 85 %-95 % of the condensate remaining after removal of water and carbon dioxide consists of hydrocarbons and their direct oxidation products. Such vapour phase pollutants during smog are very much alike on different days and in different parts of the Los Angeles Basin. They are also similar to the atmospheric pollutants during a smog in Detroit. Furthermore, the photochemical oxidation and polymerization of hydrocarbons promoted by nitrogen oxides, as disclosed by Haagen-Smit, provide apparently an adequate explanation of the formation from traffic gases of almost everything in the nature of organic impurities found in the air during a smog.

A number of other substances, probably not associated with motor fuels or motor traffic, were found in variable quantities. These included trichloroethylene, carbon tetrachloride, hydrochloric acid and sulfur dioxide. There was usually about 10 % of aromatic compounds in the condensate.

More evidence of the composition of condensable pollutants is available from a recent study of smoke and air pollution in New York-New Jersey as reported by the Interstate Sanitation Commission of New Jersey, New York and Connecticut (1958). The results are shown in Table 27. The Bayonne sample was collected downwind of an area almost exclusively occupied by petroleum refineries and storage tanks. Traffic in the area was light and did not contribute significantly to the condensate. The total pollutant concentration in the air from which the sample was obtained was about 0.5 p.p.m. The Staten Island sample was taken at the edge of a marshy area several miles downwind of an industrial area on the New Jersey mainland. The total pollutant concentration in the air at the time was about 0.03 p.p.m. Neither of the above samples showed mass patterns that resembled the normal pattern for traffic gas, although some of the pollutants

TABLE 27. ANALYSIS OF CONDENSABLE POLLUTANTS

Pollutant	Staten Island (Mole %)		Bayonne (Mole %)	
Paraffins:	48.91		67.54	
n-Butane		—		7.82
2,2-Dimethyl butane		3.60		13.71
n-Pentane		—		9.16
Propane		27.97		—
2-Methyl propane		8.78		—
2,2-Dimethyl propane		—		19.66
n-Hexane		—		8.44
3,3-Dimethyl hexane		—		1.42
n-Heptane		—		2.88
3-Ethyl heptane		—		0.34
n-Octane		—		1.04
2,4-Dimethyl pentane		8.56		—
2,4-Dimethyl-3-ethyl pentane		—		3.07
Cycloparaffins:	—		8.50	
1,1-Dimethyl cyclopentane		—		8.19
1,1,3-Trimethyl cyclohexane		—		0.31
Olefins:	11.37		16.07	
1-Butene		—		9.34
2-Methyl propene		—		2.96
3-Methyl-1-butene		—		1.50
2,3,3-Trimethyl-1-butene		0.50		—
1-Octene		8.37		1.31
1-Nonene		—		0.96
Dodecene		2.50		—
Acetylenes:	12.22		2.73	
Acetylene		9.84		2.03
1-Octyne		—		0.70
3-Hexyne		1.42		—
1-Heptyne		0.96		—
Aromatic hydrocarbons:	6.88		3.18	
Benzene		2.20		1.71
Toluene		0.86		0.81
Xylene		0.69		0.66
Trimethyl benzene		0.83		—
Diethyl benzene		0.56		—
1-Methyl-3-tert-butyl benzene		0.39		—
1,2-Diphenyl ethane		1.35		—
Chlorinated hydrocarbons:	5.45		0.18	
Chloromethane		3.91		—
Dichloroethylene		0.97		0.15
Trichloroethylene		0.57		—
Carbon tetrachloride		—		0.03
Other:	15.15		1.82	
Methyl amine		9.39		—
Diethyl amine		2.73		—
Acetic acid		3.03		—
2-Butanol		—		0.40
Sulphur dioxide		—		0.12
2,3-Dimethyl thiophene		—		0.82
3-Methyl thiophene		—		0.48

identified probably have their source in traffic gases. On the other hand, the mass spectra of samples collected on a busy street in Manhattan corresponded to a mixture very similar to normal traffic gas.

Recent developments in the separation and detection of volatile hydrocarbons and other organic compounds by gas chromatography have placed a powerful tool in the hands of workers in the field of air pollution. One method employs the thermal conductivity principle, widely used in gas analysis, for measurement after separation and fractionation of components of the mixture by adsorption followed by desorption, using an inert carrier gas. The advantages and limitations of this method are discussed by Brown & Dean (1955). Another type of detector employs a method based on flame temperature measurements during the combustion of gas fractions with hydrogen as the carrier gas (Scott, 1953).

Smog

Although the term " smog " was coined to denote smoke and fog conditions in urban and industrial areas, the composition and effects of pollution by smog gases are best known from studies on the Los Angeles problem. Severe economic losses caused by damage to vegetation, eye irritation, and rapid deterioration of rubber are attributed to the action of organic ozonides, peroxides and acids. Organic acids, aldehydes, and saturated hydrocarbons in the concentrations found in the Los Angeles smog atmosphere caused no injury to sensitive plants, such as endive, Romaine lettuce, and spinach, characteristic of the smog damage or " silverleaf " symptoms. However, a mixture of ozonides and other peroxidic compounds, acids and aldehydes formed by the interaction of ozone and olefins in low concentrations did produce smog symptoms identical with those found on crops in the field. When plants were exposed to the oxidation products of unsaturated hydrocarbons reacting with nitrogen oxides in sunlight, similar damage was produced on the leaves. It was also found that when ozone or oxides of nitrogen were combined with certain olefins in sunlight, the products were irritating to the eyes at the concentrations to be expected in the Los Angeles atmosphere. In experimental fumigations on crop plants, typical smog damage was produced by the oxidation products of ozone and straight-chain olefins of C_5 and C_6 atoms. Higher olefins of the C_7, C_8 and C_9 series were also effective in producing " silverleaf " damage on sensitive plants when converted to the corresponding ozonides and other products, but were not as active as l-pentene and l-hexene. The products of ozonization of the fraction of cracked gasoline boiling between 59°-69°C (containing C_5 and C_6 olefins) also produced typical damage on vegetation.

Similar results were obtained when plants were treated with the vapours of cracked gasoline or l-hexene, nitrogen dioxide and sunlight or ultraviolet light. Nitrogen dioxide alone, with or without ultraviolet light, or olefins alone in the presence of sunlight, did not produce these effects (Haagen-Smit, 1952; Thomas, 1951).

Haagen-Smit theory of smog formation

Of the theories that have been propounded to account for the formation of smog in the Los Angeles atmosphere by interaction of impurities with molecular oxygen, atomic oxygen and ozone, the most comprehensive and generally acceptable is that of Haagen-Smit (1952). It was found that during smogs there was a considerable increase in the concentrations of ozone and oxidant material. The concentrations reached as high as 30-40 parts per hundred million or more, much higher than in most other areas in the world where tests have been made (Bartel & Temple, 1952; Haagen-Smit, 1952). The ozone is not found in appreciable concentrations at night but only in daylight hours, beginning to form simultaneously throughout the Los Angeles basin in smoggy air shortly after dawn (Magill & Benoliel, 1952).

These facts indicate the photochemical formation of ozone or oxidant from impurities by the action of sunlight. The atmospheric studies on ozone or oxidants, oxides of nitrogen and organic pollutants such as hydrocarbons and aldehydes have been used by Haagen-Smit to establish the fundamental principles underlying the formation of smog through photochemical reactions. Thus sulfur dioxide, nitrogen dioxide, and aldehydes may absorb ultraviolet radiation in the wave-lengths present at ground level, and react in their excited states with molecular oxygen to produce atomic oxygen. Although the amount of atomic oxygen produced photochemically by aldehydes and sulfur dioxide is limited by the concentrations of these impurities, since the reactions are irreversible, this is not so for nitrogen dioxide. In the latter case, the absorption of ultraviolet light leads to the rupture of a bond to form atomic oxygen and nitric oxide. Reaction of the products with molecular oxygen leads to the formation of ozone and the regeneration of the nitrogen dioxide. The nitrogen dioxide is therefore available for repetition of the process, unless converted to nitric acid or used up in organic substitution reactions. Even low concentrations of nitrogen dioxide could produce relatively large amounts of atomic oxygen to form ozone or to react with organic pollutants to yield compounds which could cause eye irritation, crop damage, and reduced visibility. The ozone formed during smog could account for the accelerated rubber cracking, while oxidation of sulfur dioxide to the trioxide, with subsequent formation of sulfuric acid aerosol, as well as existing smoke, submicron dusts, and fumes, would still further reduce the visibility.

Chemical reactions in the atmosphere

The investigations into the Los Angeles smog problem have directed widespread attention to the unpleasant effects and economic damage that can be caused by photochemical reactions between gaseous impurities after they are released to the air. The facts cited in the Haagen-Smit theory of smog formation indicate the photochemical formation of ozone or oxidant from certain waste products by the action of sunlight. A number of photochemical reactions that could contribute to the formation of ozone in smog have been considered by Blacet (1952). These include the photochemical activation and photolysis of sulfur dioxide, aldehydes and nitrogen dioxide, all present in the Los Angeles and other city atmospheres.

The solar energy which is important photochemically near the surface of the earth is in the range of wave-length between about 8000 and 2900 Å. The intensity is relatively high above 3150 Å but drops rapidly below 3000 Å. This rules out all reactions which occur at high altitudes in the upper atmosphere, so that the only gas phase photochemical processes which can occur in the lower atmosphere must involve impurities created by human activities, if natural sources such as volcanoes and forest fires are disregarded. Substances present in the air in small amounts must have a high specific absorption in the above wave-length range in order to be of importance as a primary reactant. On the other hand, a compound present in large excess in the air might serve as a primary photochemical reactant even if its absorption were weak.

Ozone may be created in the lower atmosphere as a by-product photochemical reaction in the oxidation of sulfur dioxide to sulfuric acid in the presence of sunlight. This gas shows moderate absorption bands in the ultraviolet but the energy absorbed is insufficient to disrupt a bond in the molecule. The reaction involves the following steps if it is assumed that, initially, only activated SO_2 molecules are formed. Some of these molecules will revert to their original state but others will react with oxygen.

$$\begin{aligned} SO_2 + h\nu &= SO_2^* \\ SO_2^* + O_2 &= SO_4 \\ SO_4 + O_2 &= SO_3 + O_3 \\ H_2O + SO_3 &= H_2SO_4 \end{aligned}$$

The quantum efficiency for this process is low. Ordinarily, with high concentrations of sulfur dioxide the ozone formed would react rapidly with molecules of SO_2 to yield sulfur trioxide and oxygen. In polluted air where the SO_2 concentration is less than 1 p.p.m., the ozone may coexist with it for considerable periods or react with organic pollutants such as unsaturated hydrocarbons.

For nitrogen dioxide, Blacet gives the following photochemical reaction involving the rupture of a bond to form atomic oxygen and nitric oxide, formation of ozone by reaction of atomic oxygen with ordinary diatomic oxygen and regeneration of the nitrogen dioxide to repeat the process. The over-all process is the continued formation of ozone. Below a wave-length of 3700 Å the quantum efficiency of dissociation of nitrogen dioxide is close to unity.

$$\begin{aligned} NO_2 + h\nu &= NO + O \\ O + O_2 &= O_3 \\ NO + O_2 &= NO_3 \\ NO_3 + O_2 &= NO_2 + O_3 \\ \hline \text{In summary:} \quad 3O_2 &= 2O_3 \end{aligned}$$

In the above reactions nitrogen dioxide performs a role similar to that of a catalyst and is therefore much more effective in forming ozone than is sulfur dioxide. Cadle & Johnstone (1952) point out that nitric oxide reacts with ozone at an extremely rapid rate, much faster than the reaction between ozone and other contaminants. The following reactions can therefore take place if nitric oxide is also present in the air,

$$NO + O_3 = NO_2 + O_2$$

If ozone is present in excess, then,

$$\begin{aligned} 2\,NO_2 + O_3 &= N_2O_5 + O_2 \\ N_2O_5 + H_2O &= 2\,HNO_3 \end{aligned}$$

Thus nitric acid may be formed in the presence of water vapour.

The rate, mechanism and products of reaction of ozone with various constituents of smog have been studied for several years by the Stanford Research Institute. Ozone and olefins were found to react rapidly in the gas phase, the final products consisting of formaldehyde, higher aldehydes and polymers of unknown composition. Blacet postulates the following mechanism for the formation of ozone as a by-product in the activation of aldehydes or oxygenated organic compounds by absorption of light energy:

$$\begin{aligned} RCHO + h\nu &= RCHO^* \\ RCHO^* + O_2 &= RCO_3H \\ RCO_3H + O_2 &= RCOOH + O_3 \end{aligned}$$

Aldehydes and ketones absorb energy in the near ultraviolet sunlight with quantum yields of 0.1-0.9 to form free radicals:

$$CH_3CHO + h\nu = CH_3 + HCO$$

It is thought that the low temperature oxidation of hydrocarbons is due mostly to the free radical chain,

$$\begin{aligned} R + O_2 &= RO_2 \\ RO_2 + R^1H &= RO_2H + R^1 \end{aligned}$$

Haagen-Smit (1952) has postulated the following schematic representation of the reactions in polluted air leading to smog symptoms:

$$\mathrm{NO_2} \underset{\text{Sunlight}}{\rightleftharpoons} \mathrm{NO} + \mathrm{O} \xrightarrow{\mathrm{O_2}} \text{Oxidant}$$

$$\text{Oxidant} + \left\{\begin{array}{l} \text{Sulfur dioxide} \rightarrow \text{Sulfur trioxide} \rightarrow \text{Aerosols} \\ \text{Hydrocarbons} \rightarrow \left.\begin{array}{l}\text{Peroxides}\\ \text{Aldehydes}\\ \text{Acids}\end{array}\right\} \rightarrow \left\{\begin{array}{l}\text{Crop damage}\\ \text{Eye irritation}\\ \text{Rubber cracking}\end{array}\right.\end{array}\right.$$

According to Cadle & Johnstone (1952) and Cadle & Schadt (1952) fairly rapid reaction can occur between various olefins and ozone at concentrations that may be found in the contaminated urban atmosphere. The initial rates follow a second-order rate law although the reaction mechanisms are not known precisely and a variety of products is formed. Final products include formaldehyde, higher aldehydes, and polymers of unknown composition. Acetylene reacts with ozone at a relatively slow rate, and in city air this reaction would be of negligible consequence. Reactions of ozone with paraffin hydrocarbons and benzene were at a rate that was too small to be determined by the methods employed by Cadle. The initial second-order rate constants and half-lives of some of these reactions with ozone are shown in Table 28. Nitric oxide reacts with ozone at a rate that is about 2000 times more rapid than the reaction of l-hexene or gasoline vapour.

TABLE 28
REACTION RATES OF OZONE WITH SOME COMPOUNDS

Substance reacting with ozone	Half-life of reactants and ozone at 0.2 p.p.m. (minutes)	Half-life of reactants and ozone at 1 p.p.m. (minutes)	Rate constant p.p.m.$^{-1}$ min.$^{-1}$ at 25° C
Ethylene	1 100	220	0.0045
1-Hexene	330	66	0.015
Cyclohexene	57	12	0.087
Gasoline	380	76	0.013
Acetylene	500 000	24 000	0.0001
Nitric oxide	0.16	0.03	32.0
Nitrogen dioxide	65	13	0.077

Source: Cadle & Magill (1956). (By permission from "Air pollution handbook", by P. L. Magill, F. R. Holden & C. Ackley, editors. Copyright 1956. McGraw-Hill Book Company Inc.)

Reactions of ozone with olefins and diolefins and the photochemical decomposition of aldehydes or ketones yield organic free radicals among the products. Such reactions may occur in the contaminated urban atmosphere. Thus, the primary photochemical reaction of acetaldehyde is the rupture of the molecule to form methyl and formyl radicals. Acetone decomposes into methyl and acetyl radicals. In the presence of air, these radicals tend to react quite rapidly with oxygen to yield various products, including peroxides. For example, Mignolet (1941) has shown that diacetyl peroxide is the major product from the photochemical decomposition of acetaldehyde in the presence of oxygen. Mixtures of diacetyl and air when irradiated in sunlight have been found to produce ozone as a by-product in experiments by Haagen-Smit, Bradley & Fox (1953).

Certain other photochemical reactions that may occur in the atmosphere yield atoms or inorganic free radicals capable of acting upon saturated or unsaturated aliphatic and aromatic hydrocarbons to produce organic free radicals. Such reactive radicals and atoms include oxygen liberated in the photochemical decomposition of NO_2, chlorine atoms liberated in the action of light on molecular chlorine or nitrosyl chloride, hydrogen atoms, OH and HO_2.

The peroxy free radicals formed by the oxidation of hydrocarbon free radicals may, in turn, react further with other organic substances to yield organic peroxides and new free radicals. It is apparent, therefore, that the possibilities are almost limitless and an organic free radical may be formed in nearly every case where photochemical reactions occur in the polluted air of cities. The resultant free radical reactions can lead to polymerization and oxidation chain reactions. The chain propagating mechanism is usually terminated when the free radicals are adsorbed or destroyed by surfaces, such as airborne particulate matter, or by reaction with some substances —such as nitrogen oxides or organic compounds—that do not lead to the formation of fresh free radicals. Some of these reactions have been shown to occur in the Los Angeles atmosphere and in experimental smog chambers.

Recent Literature

The subject of the physical and chemical nature of air pollution is so broad and covers so many fields of physics and chemistry that it is almost impossible to do more than touch upon the highlights of some of the work accomplished within the last ten or fifteen years. The recent publication of the *Air pollution handbook* [1] has provided a very useful source of information, compiled by thirty-one experts in diverse fields of air pollution research and technology. There are very few journals devoted specifically to air pollution papers. Most of the scientific and technical contributions to the subject are scattered through a large number of physical, chemical, physiological, meteorological, engineering, industrial hygiene and medical publications. Some attempt has been made in the USA to provide a medium for air pollution papers in the *Journal of the Air Pollution Control Association,* but this can only serve to contain a small fraction of the material available annually for publication. The recent appearance of the *International Journal of Air Pollution* provides another excellent medium for high quality papers.

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[1] Magill, P. L., Holden, F. R. & Ackley, C., ed. (1956) *Air pollution handbook*, New York, McGraw-Hill.

Very little mention has been made in this article of the rapid progress accomplished within the last decade in methods of analysis and instrumentation. Some comprehensive reviews of this subject have been presented by Kay (1957). A Committee on Air Pollution of the American Chemical Society has organized periodic symposia on many aspects of air pollution, and within recent years the symposium has become an annual affair at national meetings of the Society. Similar committees of many other scientific societies sponsor symposia either annually or from time to time. A considerable amount of interesting information has been presented at the US Technical Conference at Washington, D.C., in May 1950 (McCabe, 1952) and at three national air pollution symposia sponsored by the Stanford Research Institute at Pasadena, California, in 1949, 1952 and 1955. Many of these papers and publications contain worth-while material on the physics and chemistry of polluted air. A recent book by Green & Lane (1957) covers the physics, physical chemistry, industrial and environmental aspects of dusts, smokes and mists in an admirable manner. Many other papers and publications, too numerous to mention here, are available to the reader interested in the subject.

Research Trends

Although much has been accomplished in the elucidation of the nature and composition of pollutants in the atmosphere, far more remains to be done. Only a limited number of the substances present in the urban environment has been identified with some degree of certainty. The large variety of substances produced in the air by oxidation, reduction, photochemical, free radical, polymerization and other reactions presents a new frontier of enormous range. Studies of the effects of contaminants on health, visibility, and on various materials used in our modern society are still in their early stages. In the meantime, methods of controlling, on a practical basis, such problems as the Los Angeles situation and many others are urgently required.

There is need for more organized research into the nature and composition or urban and industrial pollution. In the USA, the Federal Air Pollution Control Act, Public Law 159, passed in 1955, has been of invaluable assistance in providing for the conduct of research within the Public Health Service and other federal departments; for the support of research elsewhere by grants and contracts with university, state and outside agencies; and for technical aid and assistance to states and communities having special problems and also in the training of personnel for work in air pollution control. Similarly, in Great Britain, research in various aspects of air pollution has been stimulated as a result of the Report of the Committee on Air Pollution in 1954 and the passage of the Clean Air Act of 1956.

Current and near future trends in research show emphasis on the following problems, among others:

(1) The development of improved methods and techniques for the measurement, separation and identification of air contaminants.

(2) The standardization of methods of sampling and analysis of common air pollutants.

(3) The application of meteorological concepts and diffusion theory to the study of the dispersion of pollutants in the atmosphere, the formation of smog and the prediction of pollution levels.

(4) The development of improved analytical techniques, instrumentation and studies of motor vehicle exhaust gas composition under various operating conditions and the development of catalytic and other exhaust gas system control devices.

(5) The study of the action of sunlight on motor vehicle and traffic gas and of photochemical atmospheric reactions in general.

(6) The determination of the health and other effects of irradiated gaseous and vapour pollutants.

(7) The continued study of carcinogenic and other toxic substances present in the urban environment and the evaluation of their effects on health.

(8) The study of radioactive pollutants and their effects in connexion with the development of industrial uses of nuclear energy for power and transportation.

REFERENCES

Amelung, W. & Landsberg, H. (1934) *Bioklimat. Beibl.*, **1**, 49

Bartel, A. W. & Temple, J. W. (1952) *Industr. Engng Chem.*, **44**, 857

Beer, C. G. P. & Leopold, L. B. (1947) *Trans. Amer. geophys. Un.*, **28**, 173

Billington, N. S. & Saunders, D. W. (1947) *J. Inst. Heat. Vent. Engrs*, **15**, 46

Blacet, F. E. (1952) *Industr. Engng Chem.*, **44**, 1339

Brown, G. B. & Dean, R. W. (1955) *Instrument Engr*, **1**, 145

Cadle, R. D. & Johnstone, H. F. (1952) *Chemical reactions in Los Angeles smog.* In: *Proceedings of the second national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 28

Cadle, R. D. & Magill, P. L. (1956) *Chemistry of contaminated atmospheres.* In: Magill, P. L., Holden, F. R. & Ackley, C., ed. *Air pollution handbook*, New York, Section 3, p. 1

Cadle, R. D. & Schadt, C. (1952) *J. Amer. chem. Soc.*, **74**, 6002

Cartwright, J., Nagelschmidt, G. & Skidmore, F. W. (1956) *Quart. J. roy. meteorol. Soc.*, **82**, 82

Chambers, L. A., Foter, M. J. & Cholak, J. (1955) *A comparison of particulate loadings in the atmospheres of certain American cities.* In: *Proceedings of the third national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 24

Chambers, L. A., Tabor, E. C. & Foter, M. J. (1955) *A.M.A. Arch. industr. Hlth.* **16**, 17

Cholak, J. (1952) *The nature of atmospheric pollution in a number of industrial communities.* In: *Proceedings of the second national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 6

Clemo, G. R. & Miller, E. W. (1955) *Chem. & Ind.*, No. 2, 38

Cooper, R. L .(1954) *Analyst,* **79**, 573

Coste, J. H. & Wright, H. L. (1935) *Phil. Mag.*, **20**, 209

Davidson, W. F. (1942) *Monthly Weather Rec.*, **70**, 225

Dawes, J. G. (1954) *Brit. J. appl. Phys.*, **5**, 221

Dorffel, K., Lattan, H. & Rotschke, M. (1937) *Meteorol. Z.*, **54**, 16

Falk, H. L. & Kotin, P. (1956) *Chemical and biological considerations of atmospheric carcinogenic agents.* In: *Abstracts of papers, 130th meeting of the American Chemical Society, Atlantic City, N.J.*, Washington, p. 10B

Fisher, A. F. (1957) *J. Air Poll. Contr. Ass.*, **7**, 47

Gish, O. H. (1949) *Atmospheric electricity,* New York, p. 147

Great Britain, Committee on Air Pollution (1955) *Report, presented to Parliament by the Minister of Housing and Local Government, the Secretary of State for Scotland and the Minister of Fuel and Power... November 1954,* London

Great Britain, Department of Scientific and Industrial Research (1955) *The investigation of atmospheric pollution ; a report on observations in the 10 years ended 31st March, 1954,* London

Green, H. L. & Lane, W. R. (1957) *Particulate clouds : dusts, smokes and mists,* London

Greenburg, L. & Jacobs, M. B. (1956) *Industr. Engng Chem.*, **48**, 1517

Gunn, R. (1951) *Precipitation electricity.* In: American Meteorological Society, *Compendium of meteorology,* Boston, Mass., p. 128

Haagen-Smit, A. J. (1952) *Industr. Engng Chem.*, **44**, 1342

Haagen-Smit, A. J., Bradley, C. E. & Fox, M.M. (1953) *Industr. Engng Chem.*, **45**, 2086

Hagenguth, J. H. (1951) *The lightning discharge.* In: American Meteorological Society, *Compendium of meteorology,* Boston, Mass., p. 136

Hall, E. L. (1952) *Products of combustion of gaseous fuels.* In: *Proceedings of the second national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 84

Hall, S. R. (1952) *Anal. Chem.*, **24**, 996

Hemeon, W. C. L., Haines, G. F., jr & Ide, H. M. (1953) *Air repair,* **3**, 22

Hitchcock, L. B., Faith, W. L., Neiburger, M., Renzetti, N. A. & Rogers, L. H. (1955) *Air pollution situation in Los Angeles ; an aerometric survey.* In: *Proceedings of the third national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 12

Houghton, H. S. & Radford, W. H. (1938) *Physical Oceanography and Meteorography (Woods Hole Oceanographic Inst.),* 6, No. 4

International Joint Commission, Technical Advisory Board on Air Pollution (1959) *Nature and composition of pollution.* In: *Air pollution in the Detroit-Windsor Area,* Washington-Ottawa, p. 71

Johnstone, H. F. (1952) *Properties and behavior of air contaminants.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on..,* New York, p. 147

Katz, M. (1949) *Industr. Engng Chem.*, **41**, 2450

Katz, M. (1952a) *Chem. in Canada,* **4**, 157

Katz, M. (1952b) *Sources of pollution.* In: *Proceedings of the second national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 95

Katz, M. (1952c) *Amer. industr. Hyg. Ass. Quart.*, **13**, 221

Katz, M. (1954) *Recent developments in atmospheric pollution,* New York (American Gas Association, CEP-54-23. Conference, Pittsburgh, May 1954)

Katz, M. & Clayton, G. D. (1953) *Proc. Amer. Soc. Testing Materials,* **53**, 1136

Katz, M. & Cole, R. J. (1950) *Industr. Engng Chem.*, **42**, 2258

Katz, M. & Sanderson, H. P. (1958) *Filtration methods for the evaluation of aerosol contaminants.* In: American Society for Testing Materials, *Symposium on instrumentation,* Boston, Mass., p. 29

Katz, M., Sanderson, H. P. & Ferguson, M. B. (1958) *Anal. Chem.*, **30**, 1172

Kay, K. (1957) *Anal. Chem.*, **29**, 589

Kennaway, E. L. & Kennaway, N. M. (1947) *Brit. J. Cancer,* **1**, 260

King, R. A. (1950) *Industr. Engng Chem.*, **42**, 2241

Kotin, P., Falk, H. L., Mader, P. & Thomas, M. (1954) *A.M.A. Arch. industr. Hyg.*, **9**, 153

Kotin, P., Falk, H. L. & Thomas, M. (1954) *A.M.A. Arch. industr. Hyg.*, **9**, 164

Kotin, P., Falk, H. L. & Thomas, M. (1955) *A.M.A. Arch. industr. Hlth,* **11**, 113

La Mer, V. K. (1952) *Industr. Engng Chem.*, **44**, 1270

Landsberg, H. (1938) *Ergebn. kosmisch. Phys.*, **3**, 1955

Landsberg, H. E. (1951) *Meteorol. Monogr.*, **1**, No. 4, 7

Larson, G. P., Fischer, G. I. & Hamming, W. J. (1953) *Industr. Engng Chem.*, **45**, 1070

Larson, G. P., Taylor, J. R. & Hamming, W. J. (1955) *Studies of pollution levels in relation to air movement in the Los Angeles atmosphere.* In: *Proceedings of the third national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 33

Los Angeles County Air Pollution Control District (1949) *First technical and administrative report on air pollution control in Los Angeles County, 1948-49,* Los Angeles

Los Angeles County Air Pollution Control District (1951) *Second technical and administrative report on air pollution control in Los Angeles County, 1950-51,* Los Angeles

McCabe, L. C., ed. (1952) *Air pollution. Proceedings of the United States Technical Conference on...,* New York

McCabe, L. C. & Clayton, G. D. (1952) *A.M.A. Arch. industr. Hyg.*, **6**, 199

McCabe, L. C., Mader, P. P., McMahon, H. E., Hamming, W. J. & Chaney, A. L. (1949) *Industr. Engng Chem.*, **41**, 2486

Magill, P. L. & Benoliel, R. W. (1952) *Industr. Engng Chem.*, **44**, 1347

Meetham, A. R. (1952) *Atmospheric pollution,* London

Mie, G. (1908) *Ann. Phys.*, **25**, 377

Mignolet, J. (1941) *Bull. Soc. roy. Sci. Liège,* **10**, 343

Miller, C. E. (1938) *Chem. metallurg. Engng,* **45**, 132

National Smoke Abatement Society (1953) *Smokeless Air,* **23**, No. 85, 100

Neuberger, H. (1948) *Mech. Engng,* **70**, 221

New York, New Jersey & Connecticut, Interstate Sanitation Commission (1958) *Smoke and air pollution, New York-New Jersey,* New York

Proctor, B. E. (1934) *Proc. Amer. Acad. Arts & Sci.*, **69**, 315

Rayleigh (1899) *Scientific papers,* London, **1**, 92; 1903, *loc. cit.*, **4**, 400

Rodebush, W. H. (1950) *General properties of aerosols.* In: *Handbook on aerosols,* Washington (US Atomic Energy Commission), p. 60

Rogers, L. A. & Meier, F. C. (1936) *Nat. geogr. Soc. Contr. techn. Papers, Stratosphere Ser.*, **2**, 146

Rupp, W. H. (1956) *Air pollution sources and their control.* In: Magill, P. L., Holden, F. R. & Ackley, C., ed. *Air pollution handbook,* New York, Section 1, p. 1

Scott, J. A. (1953) *Publ. Hlth Rep. (Wash.)*, **68**, 474

Shaw, N. & Owens, J. S. (1925) *The smoke problems of great cities,* London

Shepherd, M., Rock, S. M., Howard, R. & Stormes, J. (1951) *Anal. Chem.*, **23**, 1431

Shore, V. & Katz, M. (1954) *Amer. industr. Hyg. Ass. Quart.*, **15**, 297

Shore, V. C. & Katz, M. (1956) *Anal. Chem.*, **28**, 1399

Silverman, L. (1951) *Chem. Engng*, **5**, 132

Sinclair, D. (1950a) *Stability of aerosols and behavior of aerosol particles.* In: *Handbook on aerosols*, Washington (US Atomic Energy Commission), p. 64

Sinclair, D. (1950b) *Optical properties of aerosols.* In: *Handbook on aerosols*, Washington (US Atomic Energy Commission), p. 81

Sinclair, D. & La Mer, V. K. (1949) *Chem. Rev.*, **44**, 345

Southern California Air Pollution Foundation (1954) *Report No. 2*, Los Angeles

Stanford Research Institute (1948-50) *The smog problem in Los Angeles County; interim report[s]*, Los Angeles

Stanford Research Institute (1954) *The smog problem in Los Angeles County*, Los Angeles

Stratton, J. A. (1931) *Electromagnetic theory*, New York

Tabor, E. C., Hauser, T. R. & Burttschell, R. H. (1956) *Characteristics of the organic particulate matter in the atmosphere of certain American cities.* In: *Abstracts of papers, 130th meeting of the American Chemical Society, Atlantic City*, Washington, p. 4B

Thomas, M. D. (1951) *Ann. Rev. Plant Physiol.*, **2**, 293

United States, Department of Health, Education, and Welfare, Public Health Service (1958) *Analysis of suspended particulate samples collected 1953-1957.* In: *Air pollution measurements of the National Air Sampling Network*, Cincinnati (Robert A. Taft Sanitary Engineering Center)

Waller, R. E. (1952) *Brit. J. Cancer*, **6**, 8

Watson, H. H. (1948) *Brit. J. industr. Med.*, **5**, 198

Weaver, E. R. & Gunther, S. (1955) *Condensable impurities in the air of Los Angeles and vicinity.* In: *Proceedings of the third national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 86

Wilson, W. S., Guenther, W. B., Lowrey, R. D. & Cain, J. C. (1952) *Trans. Amer. geophys. Un.*, **33**, 361

Harry HEIMANN, M. D. *

EFFECTS OF AIR POLLUTION ON HUMAN HEALTH

Introduction

It has long been suspected that the air we breathe is one of the environmental factors that, under some circumstances, can cause ill health, but proof of this has been lacking until relatively recently. Such proof is applicable to certain air-borne bacterial and viral infections, some allergic diseases and to poisonings due to the inhalation of certain gases and aerosols with the air we breathe. In all of these instances the air acts as a medium that permits, and may even aid in, the conveyance of the noxious agent from its source to the human host. For some of the bacterial and viral diseases and for some of the allergic conditions the evidence that the air is a carrier of the etiological agent is incontrovertible. There is less agreement among health authorities on the significance of the relationship of gases and aerosols to disease, except when these substances appear in the air of workplaces, or when cases of patent gas poisoning occur. The primary purpose of this chapter is to consider critically what is known and what is suspected about the effects upon human health of gases and aerosols when they are present in the *community* ambient air. We shall do this by considering data collected to clarify the known direct effects of these air pollutants as well as pertinent collateral information gathered from data developed for other purposes. A second object of this chapter is to recommend, as we proceed in our discussion, those areas of knowledge in which the subject of the health effects of air pollution needs further study for adequate understanding.

The air we breathe has not only life-supporting properties but also potential life-damaging properties. Under ideal conditions, the air that we must inhale has a qualitative and quantitative balance that maintains the well-being of man. When the balance among the air components is disturbed,

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health may be injured, the kind of injury and its degree being dependent upon the nature of the disturbance. Such an unhealthful imbalance may involve the physiologically necessary air gases oxygen and carbon dioxide or it may involve the so-called air pollutants. These latter are air components which are not universally or constantly present either in kind or in amount and which, in the present state of our knowledge, probably are not essential for a healthy body economy. If one were to list the constituents of ambient air that man must ordinarily breathe, besides oxygen and carbon dioxide, such a listing would include the inert gases as well as the so-called "impurities"—materials that serve no known useful function in the body. The impurities in the inhaled air do not necessarily all cause harm. Dependent upon the chemical nature of the pollutants, some may cause harm when present in the air in small concentrations and others may do so only when they reach high levels of concentration. (Table 1 lists the amounts of the relatively constant components of clean air according to one investigator, whose conclusions were based partly on his own studies and partly on a review of the reports of others. The constancy of the figures quoted is to be noted.) Except at exceedingly low levels of concentration the duration of exposure to a potentially noxious material is also of significance in relation to whether or not a given concentration will actually be harmful. When the concentration is exceedingly low the natural protective mechanism of the body may be of such effectiveness that the factor of duration of exposure is significant.

TABLE 1
RELATIVELY CONSTANT COMPONENTS
OF ATMOSPHERIC AIR

Component	Proportion expressed in %	Proportion expressed in p.p.m.
Nitrogen	78.084 ± 0.004	
Oxygen	20.946 ± 0.002	
Carbon dioxide	0.033 ± 0.001	
Argon	0.934 ± 0.001	
Neon		18.18 ± 0.040
Helium		5.24 ± 0.004
Krypton		1.14 ± 0.010
Xenon		0.087 ± 0.001
Hydrogen		0.5
Methane		2.0
Nitrous oxide (N_2O)		0.50 ± 0.1

Source: Glueckauf, E. (1951) (By permission from "Compendium of Meteorology", published in 1951 by the American Meteorological Society through support of the Geophysics Research Directorate, Air Force Cambridge Research Center, Air Research and Development Command.)

The pollution of the air that is not infrequently found in factories, workshops and mines, and the effect of such pollution upon the worker, have long been of concern to health and labour officials as well as to labour and management. This concern has led to the accumulation of a great deal of sound knowledge that can be used for the control of these unhealthful conditions. Because of the confined areas generally utilized for working-places, the concentration of potentially noxious agents tends to be higher than that which reaches outdoors from the work-place. In fact, besides flowing streams, the air is the regular medium into which waste material from work-places is discharged. That pollution which reaches the area outside the work-place is diluted by the larger volume of air that is available and, therefore, can always be expected to be at an appreciably lower level of concentration than that found inside the work-place. From occupational disease investigations we have derived some knowledge and suggestions as to how to obtain information about contaminations of the outdoor air and their possible effects upon the health of man.

On the basis of such investigations of occupational disease control problems, done in both work-place and laboratory, the safe limits of exposure to a number of known industrial air pollutants have been indicated (*A.M.A. Arch. industr. Hlth*, 1956). It is not a satisfactory procedure, however, to use these as safe-limit values when considering community air pollution. Some of the reasons for this are as follows: (*a*) potential exposure in the work-place is for about eight hours a day, whereas in the community the potential exposure continues throughout the 24 hours; (*b*) industrial workers are generally adults, in relatively good health, whereas the general public also includes the sick, and the very sensitive groups of the very young and the aged. The latter groups may be especially susceptible to adverse effects from the potentially noxious agents; (*c*) intermittent exposures such as occur in industry allow the organism to recover from injury during the period of time when there is no exposure, whereas continuous exposure, such as that which may occur with community air pollution, lessens this opportunity for recovery; and (*d*) inside the work-place atmospheric conditions are subject to control, whereas the weather outside is generally uncontrollable and may be such that it either alters the air contaminants themselves or modifies their biological effects.

An example of chemical alteration of air pollutants by weather is that which occurs in Los Angeles, wherein the action of the sun's rays upon the air-contained oxides of nitrogen and olefinic hydrocarbons results in the appearance of ozone and nitro-olefins in the air. An example of the modification by weather of the biological effect of air pollutants is the simple one of dilution that occurs through the turbulence within the air mass. A second more complicated example is that involved in recent studies which showed that experimental animals challenged with known toxic agents respond more forcefully as the ambient temperature is raised (Baetjer & Smith, 1956).

Not only is there an influence of weather and climate on the biological effects of air pollutants, but it has long been believed that weather and climate *per se* also appear to have significant effects on the health of man. So strong has this belief been that the health professions have long considered certain geographical locations more salubrious than others because of their weather. If living in such places is in fact beneficial for persons afflicted with certain diseases, it is not now certain that this effect is always directly attributable to the weather and climate: it may be due to the influence that local weather and climate have on the air-contained materials—for example, on the accumulation or the chemical kinetics of air pollutants. The significance of the weather factor in acute air pollution episodes, as is discussed below, is well known, whereas its importance in relation to long-continued exposure to low levels of air pollutants is only now being estimated. The relationship of the weather and many other factors to the presence of positive and negative ions in the air and, in turn, the effect of such ions on health, is poorly understood at present. Finally, the influence that air pollution may have on weather itself may be significant on occasion (Gunn & Phillips, 1957).

The sources of air pollution are ubiquitous. Naturally occurring pollutants, over which man has either very little or no control, originate from forest fires started by lightning, volcanic eruptions, organic materials emanating from plants, decaying vegetation, dust storms and salt from sea spray. Man-made air pollution has as its principal source combustion for the production of energy. This combustion may take place in the home, in the work-place, or in mechanical transportation equipment. A source that is generally of secondary importance, but one that at times turns out to be of great significance in air pollution problems, is that involving vaporization of liquids or pulverization of solid materials, generally for industrial purposes. It is emphasized that industry is not the only source of community man-made air pollution, although in some places it is the major one. Home-heating, for example, was concluded to be the major source in the London air pollution episode of 1952, which is described below. Even the very essential process of food-cooking, especially at the high temperatures required for frying, possibly contributes harmful pollutants, at present unsuspected, to the air in the home.

Acute Air Pollution Episodes

With the outbreaks of sickness and death associated with air pollution, and ascribed to the pollution in some instances, the health professions began to show genuine interest in the fact that air contaminants could cause harm to man. The outbreak in the Meuse Valley of Belgium in 1930, the 1948 epidemic in the City of Donora, Pennsylvania, in the USA, and the London episode of December 1952, directed attention to this phenomenon. Following studies and reports of those incidents it was generally accepted that air pollution was an undesirable condition that could have serious health consequences. Each of the known episodes that have occurred will now be summarized briefly, and emphasis will be placed on what we may learn from them for use in the future.

Meuse Valley, 1930

During the first week of December 1930, all of Belgium was blanketed by fog which was due in part to anticyclonic weather conditions (Alexander, 1931; Batta, Firket & Leclerc, 1933; *Brit. med. J.*, 1931; Firket, 1931, 1936; Haldane, 1931; *J. Amer. med. Ass.*, 1937; van Leeuwen, 1931, 1933; Mage & Batta, 1932a, b; Roholm, 1936, 1938). As would be expected, the situation was especially marked in the river valleys. The Valley of the Meuse River, extending from Huy to Seraing, which was one of these fog-bound areas, was affected, in addition, by a temperature inversion. Along a 15-mile [1] length of the Valley, from which the hills on either side rise to a height of 250-350 feet [2] above the river banks, were located many heavy industrial plants, including coke ovens, blast furnaces, steel mills, power plants, glass factories, lime furnaces, zinc reduction plants, a sulfuric acid plant, and an artificial fertilizer plant (Batta et al., 1933).

Beginning with the third day of the weather abnormality people in this portion of the Valley became ill with respiratory tract complaints, and some 60 of them died. The exact number who were ill was never determined, although it was estimated as having been in the thousands. The number of deaths was considered as having been about 10.5 times the expected amount for an equivalent period of time and season under ordinary circumstances. The illness affected persons of all ages and both sexes, consisting of an irritation of the membranes of the body that are exposed to the air, especially those of the respiratory tract. The malady was manifested by the following symptoms: epiphora, throat irritation, hoarseness, cough, shortness of breath, a sense of constriction of the chest, nausea and some vomiting. Cough, both productive and non-productive, as well shortness of breath, was the dominant symptom. Treatment, effective in some of the cases, was essentially symptomatic, consisting of antispasmodic drugs to relieve dyspnoea. The persons who died were the elderly and those who had chronic disease of the heart or of the lungs. Autopsies confirmed the conclusions reached from studies carried out on the living, which indicated that the probable causative agent was an irritant chemical substance

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[1] 1 mile = 1.6 km.

[2] 1 ft = 0.3 m.

(or group of substances) that acted upon the lining of the respiratory tract. The organs and tissues that were examined showed no effects that could be ascribable to a toxic agent affecting parts of the body other than the respiratory tract.

With the termination of the fog no new cases of illness appeared, although a few deaths occurred among those who had become very ill during the fog. There is some suggestion in the reports of the episode that new cases of illness stopped appearing even before the fog lifted.

A search, beginning almost immediately after the fog lifted, was made to discover, if possible, the specific causative factor responsible for the illnesses and deaths. It was clearly shown that besides the freak weather nothing unusual was going on in the area—i.e., none of the factories had been operating in an abnormal way. After careful consideration of all the gases and aerosols that were discharged into the air of this industrial valley it was believed that only an irritant air pollutant could have caused the illnesses and deaths, and that it was probably a mixture of the sulfur oxides, sulfur dioxide gas and sulfur trioxide aerosol. It was also considered that other aerosols and irritant gases may have contributed to the chemical changes of those irritants and to the clinical conditions, but in a small degree. Thus, it was felt that the air-borne oxides of nitrogen may have aided in the change of the sulfur dioxide to sulfur trioxide, and that the presence in the air of some of the solid particulate metal oxides also may have aided in the chemical reaction, some even acting as catalysts. Finally, it was felt that aerosols ordinarily considered as inert physiologically may have played a role in the illnesses by conveying the irritant gases deeper into the lungs than the gases might have gone without the presence of these air-borne aerosols to which the gas molecules could attach themselves.

Consideration was given to the possibility that air-borne irritant fluoride compounds were responsible for the human effects. This, however, was carefully ruled out by the official investigators of the episode. A few years later the possibility that the fluoride gases were an etiological factor was again mentioned (*J. Amer. med. Ass.*, 1937; van Leeuwen, 1933; Mage & Batta, 1932a; Roholm, 1936, 1938).

The role that weather played in the episode was felt to be a significant one, for it permitted the accumulation to toxic concentrations of those pollutants normally discharged into the air by the factories in the Valley. Careful review of the weather for years prior to the one in which the episode occurred showed that the area in question had had similar weather abnormalities previously, but that most of them had lasted for periods of time that were shorter than the one in 1930 and that they had not been associated with such serious health effects as those found in 1930. For a comparable weather episode in the year 1911, for example, illness records appeared to show a similarity to those of the episode of 1930, except that there were no deaths ascribed to the earlier incident (Batta et al., 1933).

Donora, 1948

During the last week of October 1948, a temperature inversion and an anticyclonic weather condition characterized by little or no air movement occurred over a wide area of the north-eastern portion of the USA (Fletcher, 1949; Schrenk et al., 1949; Willett, quoted by Schrenk et al., 1949). The unusually prolonged stable atmospheric condition, associated with fog, resulted in the accumulation of atmospheric contaminants to abnormal levels of concentration, particularly in highly industrialized areas. The city of Donora was one such area.

Donora is located on the inside of a sharp horseshoe bend of the meandering Monongahela River some 30 miles south of Pittsburgh. The narrow plain immediately adjacent to the river is occupied on the Donora side of the river by a large steel and a large zinc reduction plant. Just beyond this narrow plain, as well as the one in the town of Webster on the opposite bank, the terrain rises sharply. On the Donora side the hills rapidly reach a height of 400 feet (1150 feet above sea level) in a 10% grade.

On the morning of 27 October 1948, the atmosphere of Donora became very still, there was a marked weather inversion and fog enveloped the city. This weather phenomenon was associated with an accumulation of atmospheric contaminants. The condition continued until 31 October, when an afternoon rain occurred and the change of weather was associated with clarification of the air. In that interval of four days there was a great deal of respiratory tract illness among the residents. During that same period there were 17 deaths in the community, which normally experienced an average of two deaths in an equivalent period of time[1] (Fletcher, 1949; McCabe, 1952; Mills, 1949a,b, 1950; Roueché, 1950; Schrenk et al., 1949; Shilen, 1949; Shilen et al., 1949). (See Table 2.)

The US Public Health Service was asked to assist the state health authorities to make an epidemiological study of the episode, with the ultimate object of avoiding such occurrences in the future (Schrenk et al., 1949). The study was begun early in December of the same year, about two months after the episode. The investigation was planned to determine the following: (*a*) the health effects that occurred among the people and domestic animals; (*b*) the qualitative and quantitative nature of the contaminants that had accumulated in the air; and (*c*) the meteorological conditions that existed at the time of the episode.

The primary method used to study the human effects was the household canvass by questionnaire to obtain data on descriptions of the sicknesses that had occurred and of their prevalence. This canvass was carried out by public health nurses in a statistically valid sample of the population. (One-third of the total population constituted the sample.) For a selected

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[1] During the days immediately following the episode three more deaths occurred among persons who became ill during the episode.

TABLE 2

CRUDE MORTALITY DATA FOR THE BOROUGH OF DONORA
FOR SEVEN SUCCESSIVE TWO-WEEK PERIODS,
12 SEPTEMBER-18 DECEMBER, 1948,
INCLUDING THE PERIOD OF THE AIR POLLUTION EPISODE OF OCTOBER 1948

Two-week period	No. of deaths	Deaths per 1000 persons in population (12 300, approximately)	Difference from average of the seven two-week periods (0.407 per 1000)	
			Rate	%
12 Sept.-25 Sept.	2	0.163	—0.244	—59.9
26 Sept.-9 Oct.	3	0.244	—0.163	—40.0
10 Oct.-23 Oct.	1	0.081	—0.326	—80.1
24 Oct.-6 Nov. [a]	16 [b]	1.301	+0.894	+219.7 [c]
7 Nov.-20 Nov.	7	0.569	+0.162	+39.8
21 Nov.-4 Dec.	4	0.325	—0.082	—20.1
5 Dec.-18 Dec.	2	0.163	—0.244	—59.9
Average		0.407		

Source: Schrenk et al. (1949), p. 74, Table 36.

[a] Includes the period of the air pollution episode.

[b] The Borough of Donora per se had 16 deaths during the two-week interval. The mortality data noted in other sections of this report refer to that of the Donora area which includes the Borough of Donora, Carroll Township and Webster, for purposes of this report.

[c] If one excludes the period of 24 October-6 November from the average of the death occurrences for the two-week periods, it will be found that the mortality rate during the period of 24 October-6 November comes to about 530% i.e., it is five times this new average. The severe test that we applied in the preparation of the table shows only a trebling of the average for the two-week period that included the episode.

portion of whose who were said to have been ill, physicians obtained more detailed clinical information by interview. Further medical data were obtained from a review of the medical records of patients who had been hospitalized during the episode and from autopsies. Blood studies were made for three purposes: (*a*) to determine the blood content of influenza antibodies, since the outbreak of illnesses in the Donora episode resembled in some respects that of an epidemic of influenza; (*b*) to enumerate the percentage of polymorphonuclear eosinophilic leukocytes in the blood, to be used as an indicator of the part that allergy may have played in the clinical manifestations; and (*c*) to evaluate the presence in the blood of abnormal chemical elements, either in kind or in amount. Chest roentgenograms were made of a limited number of persons. Teeth, bones and urine samples were chemically examined for fluoride content to provide auxiliary evidence in regard to the relationship that fluorides may have had to the acute clinical manifestations. A veterinary physician investigated the occurrence of sickness among both farm animals and pets during the episode. Finally, records of general morbidity and mortality of the community were studied for varying periods of time before the episode.

The aerometric and other engineering studies were also done after the episode was over. The engineers attempted to determine the nature and amounts of the materials which were present in the ambient air during the episode. Their studies included a period of time when the local factories were operating fully during a later short weather inversion. The meteorologists made a study of local weather conditions, reviewing whatever past weather records were available.

From a review of the biological studies, correlated with the engineering and meteorological data, it was believed that some definite conclusions were warranted. These conclusions are noted in the material that follows.

During the episode 5910 persons (42.7 % of the population [1]) were affected by symptoms of illness. The affection from which they suffered was initially an irritation of the respiratory tract, eyes, nose, and throat; this affection varied in degree from very " mild " to very " severe ". The classification as to the severity of the illness of the affected persons was based upon the number and kind of symptoms that were reported for them, the duration of disability if such disability occurred, the need for medical care as determined by the attempt having been made to obtain the services of a physician, whether or not the attempt was successful, and the outcome of the illness. The symptoms were also classified in three grades—namely, mild, moderate and severe. The following were considered as mild symptoms for this purpose: smarting of the eyes, lacrimation, nasal discharge, a sense of constriction of the throat, soreness of the throat, non-productive cough, nausea without vomiting, headache, weakness and muscular aches and pains. The symptoms considered as moderate were productive cough, a sense of tightness or feeling of pressure in the anterior chest, dyspnoea, vomiting and diarrhoea. The single symptom considered as severe was orthopnoea. Based upon these symptoms and the other pertinent data noted above, the scheme for the classification was arbitrarily developed as shown in Table 3.

Cough was the single dominant symptom. After cough, the frequency of other symptoms was in the following descending order: soreness of throat, a sense of chest constriction, headache, dyspnoea, a burning sensation of the eyes, orthopnoea, lacrimation, vomiting, nausea without vomiting and excessive nasal discharge (Table 4). In 15.5 % of the population the symptoms were classed as " mild "; in 16.8 % they were " moderate ", and in 10.4 % the symptom complex was described as involving " severe " affection. Neither the prevalence nor the severity of illness was influenced by sex, race, occupational status, length of residence in the community, or degree of physical activity at the time that the symptoms began. There was, however, a relationship between both frequency and severity of illness and

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[1] The data on illness incidence were based on the population of the Donora " area ", estimated at 13 839 persons, and included 12 927 for Donora and adjoining Carroll township and 912 for the town of Webster.

TABLE 3

CLASSIFICATION OF SEVERITY OF ILLNESS PRODUCED BY AIR POLLUTION IN DONORA

Number and kind of symptoms	Additional factors for classifying degree or severity of illness		
	No period of disability; no medical care needed; no symptoms still present at time of survey	Disability of 1-3 days; and/or medical care needed; and/or symptoms still present at time of survey	Disability of 4 days or longer
1 to 3 “ mild ” symptoms	Mild	Moderate	Moderate
4 or more “ mild ” symptoms	Moderate	Moderate	Severe
1 to 3 “ moderate ” symptoms with or without “ mild ” symptoms	Moderate	Moderate	Severe
4 or more “ moderate ” symptoms with or without “ mild ” symptoms	Moderate	Severe	Severe
Orthopnoea with or without other symptoms	Moderate	Severe	Severe

increasing age. Thus, whereas the sickness frequency in the total population was a little over 40 %, over 60 % of those who were 65 years of age or more reported some degree of illness, and about one-half of the sick in this older age-group were severely ill (Table 5). The illnesses of the largest number of persons began on the third day of the episode (Fig. 1).

Of the 17 deaths that occurred in the Donora area during the episode, 15 occurred on the third day of the incident. The ages of the persons who died ranged from 52 to 84 years, with a mean of 65 years. Neither principal past employment, duration of residence in the community, nor sex was epidemiologically significant in the occurrence of the fatal illnesses. Only in the severity and in the outcome did the fatal cases differ from the very ill persons who did not die. Pre-existing disease of the cardio-respiratory system appeared as the single significant common factor among the fatally ill. (In four of the 17 fatal cases no history of chronic or acute disease prior to the episode was obtained.)

Investigation of the blood for specific antibodies made by the haemagglutination-inhibition method of Salk (1944) for influenza B, A and A1 resulted in titres that led to the conclusion that epidemic influenza did not play a part in the outbreak of illness.

Studies for frequency of dental caries and dental fluorosis, urinary excretion of fluoride, and fluoride content of animal bones indicated that the ambient air of Donora ordinarily contained a very low concentration of

TABLE 4

SYMPTOMS PRODUCED BY AIR POLLUTION IN DONORA, IN DECREASING ORDER OF OCCURRENCE IN ALL AGES

Symptom	%
Cough	33.1
Non-productive	20.2
Productive	12.9
Sore throat	23.1
Constriction of the chest	21.5
Headache	17.0
Dyspnoea without orthopnoea	12.9
Smarting of the eyes	12.3
Orthopnoea	8.4
Lacrimation	8.0
Vomiting	7.4
Nausea without vomiting	7.1
Nasal discharge	6.6
Fever	2.6
Choking	2.3
Aches and pains	1.9
Weakness	1.8
Cyanosis	1.0
Diarrhoea	0.1

Source: Ashe (1952). (By permission from "United States Technical Conference on Air Pollution" by L. C. McCabe, editor. Copyright 1952. McGraw-Hill Book Company, Inc.)

fluorides when compared with similar communities in the USA. This was considered as *indirect* evidence that an accumulation of fluorides to noxious levels during the episode was very unlikely to have occurred. For this reason it was felt that irritant fluoride inhalation could not have accounted for the illnesses or deaths.

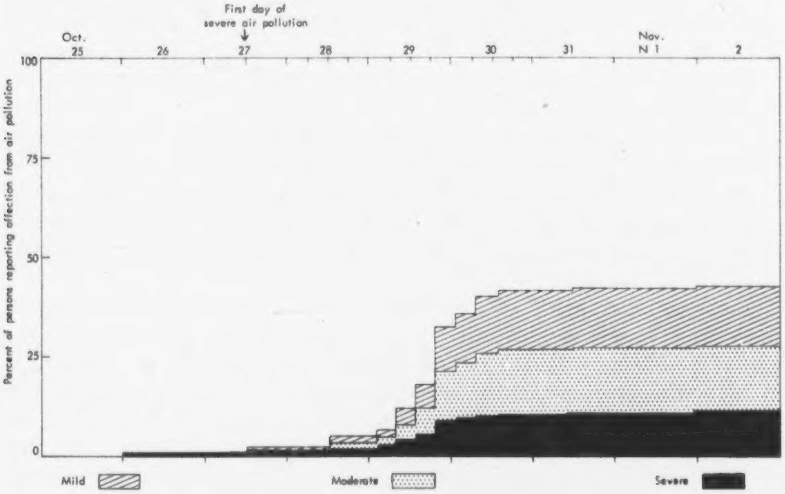
The tests made indicated in total that, since during ordinary times fluorides in the Donora air were present in exceedingly minimal amounts, if at all, then it is not reasonable to expect that such exceedingly small amounts could have accumulated during the abnormal weather period to reach a level of concentration that would be significant as a respiratory-tract irritant.

Analyses were made of morbidity records, available only for Donora residents who were employed in the local factories, and of the community's mortality records. These analyses showed that, with only two exceptions in relation to time, neither the health patterns nor the mortality patterns of the people of Donora were essentially different from those of people in nearby communities that were comparable socio-economically. The first of these exceptions was the obvious one of the period covering the acute

TABLE 5
INCIDENCE OF ILLNESS AMONG PERSONS OF INDICATED AGE IN DONORA AREA

Age in years	% of persons of indicated age reporting affection from air pollution			
	Total	Degree of affection		
		Mild	Moderate	Severe
All ages	42.7	15.5	16.8	10.4
Under 6	15.9	9.8	4.3	1.8
6-12	29.6	15.7	11.0	2.9
13-19	27.3	15.2	9.6	2.5
20-24	31.2	13.9	13.1	4.2
25-29	40.3	18.8	14.9	6.6
30-34	48.0	17.1	22.8	8.1
35-39	52.3	19.5	23.7	9.1
40-44	54.9	22.2	21.8	10.9
45-49	57.1	17.8	24.7	14.6
50-54	59.4	18.9	22.0	18.5
55-59	58.1	13.7	24.0	20.4
60-64	63.3	11.0	22.9	29.4
65 and over	59.8	10.6	20.3	28.9

FIG. 1
CUMULATIVE INCIDENCE OF AIR POLLUTION AFFECTION IN THE DONORA AREA, 25 OCTOBER TO 2 NOVEMBER 1948



episode of October 1948. The second was that mortality records revealed that a crisis had occurred in Donora in 1945 similar to the 1948 episode. It appeared that in April of 1945 the death rate of the city was higher than for any other month during 1945-48. Findings in regard to atmospheric conditions, resembling those of October 1948, suggested that the environment may have contributed to the higher incidence of deaths due to cardiovascular disease during that time.

In the October 1948 episode autopsies were carried out on only five of the bodies of the persons who died. Three were of fatalities that occurred during the episode, and two of persons who died later. Of these, three showed acute irritative changes in the lungs, characterized by capillary dilatation, haemorrhage, oedema, purulent bronchitis and purulent bronchiolitis. Chronic cardiovascular disease, the origin of which antedated the episode, was a prominent finding in the autopsy examinations. This observation confirmed the conclusion, previously reached on clinical grounds, regarding the significance of pre-existing heart disease in the nature of the illness that developed during the episode.

Correlation of the engineering and meteorological data with the medical findings, for the purpose of determining the specific substance or substances which might have served as the immediate cause of the illnesses and deaths, led to an impasse. From these sets of data, developed from information collected after the episode, it was not possible to conclude with certainty that any one of the measured air-borne materials that were estimated or actually measured could have accounted for the syndrome. It appeared reasonable, however, to conclude that the health effects were probably due to a combination of materials. We concluded that no single substance was responsible, but that the illness was probably due to a combination of the action of two or more of the air contaminants. It appeared that the culprit was a combination of sulfur dioxide (estimated to have reached a level of from 0.5 p.p.m. to 2.0 p.p.m.) (Hemeon, 1955) together with its oxidation products and non-specific particulate matter (Schrenk et al., 1949). It was not possible to determine the significance of other potential irritants, although, in the light of existing knowledge, we doubted that they were significant.

On the basis of the reported data, and additional sources of information collected later, subsequent to our report other workers in the field of air pollution studies continued to discuss possible immediate causes of the illnesses. One suggested that solid air-borne materials, specifically " metal " ammonium sulfates, derived from the reaction of the sulfur oxides with metallic elements and compounds, constituted the likely cause of the health effects (Hemeon, 1955). A second investigator suggested that the oxides of nitrogen constituted the culpable agent (Mills, 1950). Another believed that the accumulation in the air of irritant fluoride compounds was responsible (*Chem. Engng News*, 1948). The data in the official report did not support the thesis that either the oxides of nitrogen or irritating fluoride compounds could have accounted for the sicknesses. The possibility of the effects having been caused by " metal " ammonium sulfates, it seems to us now, requires serious consideration.

London, 1952

From 5 to 9 December 1952, many parts of the British Isles were enveloped in a fog associated with anticyclonic weather conditions and a temperature inversion. The broad Thames Valley, and especially London, was severly involved in this weather abnormality, which was associated with an unusual frequency of illness and death that went unrecognized substantially until after its occurrence. Although such a weather peculiarity associated with increase of sickness had occurred previously in the region, the unusually severe episode of December 1952 was well documented and is therefore selected for presentation in this review (Abercrombie, 1953; *Brit. med. J.*, 1953; Douglas & Stewart, 1953; England and Wales, Ministry of Health, 1954; Fry, 1953; Great Britain, Committee on Air Pollution, 1953, 1955; Lawther, 1955; Logan, 1953; McCabe, 1954; Scott, 1953; Smithard, 1954; Walpole, cited by Cullumbine, Pattle & Burgess, 1955; Wilkins, 1954).

Within about 12 hours after the heavy fog began and the air pollution accumulation started, an inordinately large number of people in Greater London began to become ill with respiratory tract symptoms. During the episode an unusually large number of deaths occurred (England and Wales, Ministry of Health, 1954; Fry, 1953). The symptoms of the illnesses consisted of cough with relatively little sputum, nasal discharge, sore throat and sudden attacks of vomiting. The more severely ill were those who " gave a history of previous chest trouble... i.e., chronic bronchitis, asthma, bronchiectasis, or one of the forms of pulmonary fibrosis " (Fry, 1953). The illnesses generally had a sudden onset, many of the more severe cases beginning on the third or fourth day of the fog. From the very ill patients the following signs were elicited, in addition to the symptoms noted above: dyspnoea, cyanosis, moderate fever and râles and rhonchi indicating bronchospasm. There was a fair response of the symptoms to antispasmodic drugs in some of the cases. The illness appeared to be modified by sex and age, for men were attacked with greater frequency than women and most of the sick were over 45 years of age.

Hospitals had an appreciably higher number of admissions of new patients than was normal for such a period of the year. The increase was particularly high for cases of acute respiratory tract disease, which " nearly quadrupled " for one institution (Abercrombie, 1953). Heart disease admissions to hospitals also increased sharply.

Although the incidence of illness could not be estimated with any degree of certainty (England and Wales, Ministry of Health, 1954), mortality incidence was available through the Registrar General's records; these death records were carefully studied and analysed, and a summary of the data was reported. From the incomplete morbidity records available, it was ajudged that although morbidity rates increased, the increase was not in an expected proportion to that of mortality. The mortality records showed that for a two-week period that covered the week of the episode and the week after there was a total of about 4000 excess deaths in Greater London, compared with a similar period in previous years, and it was felt that this excess of deaths was " brought about by the fog incident " (Logan, 1953). The increase in death frequency began on the first day of the episode. During the period in question other large towns in England showed only a moderate rise in the number of deaths compared to that of London, and the Central London area showed a larger increase than did the " outer ring " of the city.

" All ages shared to some extent in the increased mortality and . . . it was by no means confined to the very young or the very old " (Logan, 1953). The older persons (45 years of age and over), however, had the highest increment, and the very young (under 1 year of age) also had an unusually high frequency of deaths (Table 6). The bulk of the people who died were recorded as having had chronic bronchitis, bronchopneumonia, other lung disease, or disease of the heart (Table 7). Chronic bronchitis was especially prominent as a pre-existing disease in the fatal cases.

Correlations were made between the incidence of both sickness and death in the London episode and the air-borne quantities of smoke and of sulfur dioxide, up to 60 % of which was believed to have been due to domestic fires. Smoke concentrations during the fog were found to be five times

TABLE 6

REGISTERED DEATHS IN LONDON ADMINISTRATIVE COUNTY, BY AGE; COMPARISON OF SEVEN-DAY PERIOD BEFORE THE 1952 EPISODE WITH THE SEVEN-DAY PERIOD THAT INCLUDED THE EPISODE

Age	7-day period preceding the episode (A)	7-day period that included the episode (B)	Ratio of (B) to (A)
All ages	945	2484	2.6
Under 4 weeks	16	28	1.8
4 weeks to 1 year	12	26	2.2
1-14 years	10	13	1.3
15-44 years	61	99	1.6
45-64 years	237	652	2.8
65-74 years	254	717	2.8
75 years and over	355	949	2.7

Source: Logan (1953).

TABLE 7

NUMBER AND RATIO OF DEATHS IN LONDON ADMINISTRATIVE COUNTY BY SELECTED CAUSES FOR SEVEN-DAY PERIOD PRECEDING THE 1952 EPISODE, THE SEVEN-DAY PERIOD THAT INCLUDED THE EPISODE, AND THE SEVEN-DAY PERIOD FOLLOWING THE EPISODE

Cause of death	7 days before episode (A)	7-day period including period of episode (B)	Ratio of (B) to (A)	7-day period after episode (C)	Ratio of (C) to (A)
Respiratory tuberculosis	14	77	5.5	37	2.6
Cancer of the lung	45	69	1.5	32	0.7
Pneumonia (excluding those under 1 year of age)	45	168	3.7	125	2.8
Bronchitis	76	704	9.3	396	5.2
Influenza	2	24	12.0	9	4.5
Other respiratory disease	9	52	5.8	21	2.3
Coronary heart disease	118	281	2.4	152	1.3
Myocardial degeneration	88	244	2.8	131	1.5
Suicide	10	10	1.0	7	0.7

Source: Logan (1953).

greater than those found at other times. The sulfur dioxide level rose to 1.3 at one location for one short period of time. The general average air concentration of sulfur dioxide during the episode was 0.7 p.p.m.—about six times the usual level. Measurements of air concentrations of sulfur dioxide did not show levels that are currently generally considered toxic (Amdur, Melvin & Drinker, 1953; Amdur, Silverman & Drinker, 1952). Authorities studying the London episode considered the sulfur dioxide level only as one index of pollution. It was not known then, nor is it now, exactly what substances in the air should be measured in such episodes as the definite culpable toxic agent.

Careful consideration was given to all the factors that might have been responsible for the sicknesses and deaths that occurred during the period. The ones that received most credence were the fog, the prevailing low temperature, the sulfur oxides and the solid matter in coal smoke. The subject of specific etiology was summed up in this manner in a government report: " While the available evidence does not allow of a clear indictment of any one constituent of the fog, the conclusion is inescapable that the excessive mortality and increased incidence of respiratory illness during and immediately after the fog of December 1952 were the result of irritation of the respiratory tract by contaminants of the fog. The irritants mainly responsible were probably those derived from the combustion of coal and its products and their lethal effects were almost wholly exercised in persons already suffering from chronic respiratory or cardiovascular disorders" (Great Britain, Committee on Air Pollution, 1953, 1955).

The coal and coke that is used in Great Britain has an average sulfur content of 1.5%. The manner of burning this solid fuel in relatively inefficient open grates is probably also of significance in its contribution of sulfur gases to the ambient air (Parker, 1950). It was estimated that in 1948, for example, there were 190 million long tons [1] of coal used in Great Britain and this accounted for 4.7 million tons of sulfur dioxide discharged into the air. This was divided by source as shown in Table 8.

TABLE 8

MAIN USES OF COAL AND ESTIMATES OF RESULTANT SULFUR DIOXIDE POLLUTION IN GREAT BRITAIN FOR 1948

Type and use of fuel	Estimated annual pollution by sulfur dioxide discharged from fuel combustion	
	Amount in millions of long tons *a*	%
Coal		
Domestic use	0.9	19.1
Electricity-generating stations	0.8	17.0
Railways	0.4	8.5
Other industrial uses	1.7	36.2
Coke and gas		
Coke ovens and coke use	0.5	10.6
Gas industry		
At gasworks	0.1	2.1
Use of gas	small	small
Use of coke	0.3	6.4
Total	4.7	99.9

Source: Isaac (1953), p. 213.

a 1 long ton = 1.016 metric tons.

Similar air pollution episodes, but possibly of a less violent nature, had happened in London before the 1952 crisis. Prior to that crisis, a major increase in mortality was associated with the following periods of severe fog: December 1873, January 1880, February 1882, December 1891, December 1892, and November 1948 (Brodie, 1891; Logan, 1956; Parker, 1957; Russell, 1924). In the 1948 episode there were 300 excess deaths

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[1] 1 long ton = 1.016 metric tons.

(Logan, 1949). A later episode, in the winter of 1956, accounted for 1000 excess deaths. One British report (Logan, 1956) mentioned that although there was an apparent infrequency of such incidents in the 19th century and the first half of the 20th, the appearance of three such incidents within the eight-year period prior to 1957 " was disquieting ". The reasons for the increased occurrence of such episodes are obscure at this time (Bourne & Rushin, 1950; Brodie, 1905; Douglas & Stewart, 1953; Marshall, 1952).

The 1952 episode was severe enough to incite public opinion to the degree that the Government called for an investigation. This resulted in a series of recommendations aimed at preventing recurrences (Great Britain, Committee on Air Pollution, 1953, 1955).

Los Angeles

The County of Los Angeles, which includes within its bounderies the City of Los Angeles, is located partly along the narrow Pacific coastal plain in the southern portion of the State of California. The land rises gradually eastward from the coastline for a distance of about 30 miles, climbing approximately 2000 feet until it abuts against the mountains. There are also mountains to the north and south of the area of the county included in the sloping plain. The natural land barriers, together with the light breeze from the ocean side, form a pocket in which the air moves relatively sluggishly in a horizontal direction. In this area of 1600 square miles [1] live approximately five million people of whom half are congregated in the City of Los Angeles.

For some time this region has been affected during the summer and early autumn by occurrences of relatively high concentrations of air pollution—locally termed " smog " [2]—that cause irritation of the eyes, nose and throat, damage to vegetation, and haziness of the atmosphere (Abrams, 1949; Cadle & Magill, 1951; California State Department of Public Health, 1955, 1956; Faith, 1954; Kennedy, 1954; Magill, 1949; Senn, 1948; Stanford Research Institute, 1948, 1949, 1950, 1954).

Preliminary medical investigations in Los Angeles indicate that during periods of smog there is a large number of complaints of eye irritation and some irritation of the nose and throat, but the rates of occurrence of these subjective complaints have not been adequately recorded. The symptoms disappear when the air pollution fades, and it seems that they leave no subjective or objective residue. An interesting investigation, still in progress in Los Angeles, involves the medical observation of a selected group of elderly persons to determine whether or not frequency of hospitalization or death in this group is associated with higher levels of air pollution. Initial results indicate that there is no such relationship even though this group is very sensitive to " stress, " as is shown by their poor adaptation to sharp elevations of the temperature of the ambient air. According to local surveys general sickness incidence and air pollution in Los Angeles show no obvious relationship. Hospital admissions for respiratory and cardiac diseases show no abnormality during periods of smog. It would appear that children in school become more unruly during days of high air pollution. These and other studies of the possible relationship of human health to smog in Los Angeles are continuing.

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[1] 1 sq. mile = 2.6 km^2.

[2] According to Webster's *New international dictionary of the English language*, 2nd ed., 1956, Springfield, Ill., smog is " a fog made heavier and darker by the smoke of a city ". The word is a blend of *smoke* and *fog*. It is decidedly a misnomer in this instance since the condition to which it refers in Los Angeles is due neither to fog nor to smoke. In fact, it is said that when either fog or smoke is present in the air over the city, " smog " does not occur.

The specific agents possibly responsible for the smog are under intensive study in Los Angeles and in laboratories scattered over the entire country. The following is a list of air-borne substances, and groups of substances, that have been or are currently considered as causally related to the smog: (1) sulfur oxides (sulfur dioxide and sulfur trioxide), (2) oxides of nitrogen, (3) aldehydes, (4) ketones, (5) acids, (6) chlorinated hydrocarbons, (7) acrolein (or an " acrolein-like " substance), (8) chlorinated aldehydes (and related substances such as chloracetone, trichloraceticaldehyde, monochloracetic acid, alphachlorproprionic acid, and chloracetophenone), (9) formyl compounds (formic acid and formaldehyde), (10) ozonides and peroxides of hydrocarbons of the series present in gasoline, especially the straight-chain hydrocarbons containing double bonds, (11) ozone, (12) nitro-olefins, (13) peracylnitrites, (14) organic free radicals, and (15) carbon suboxide.

It is considered that the gases and aerosols that are discharged into the air may not be irritant *per se*. What is more likely is that the substances which normally reach the air are acted upon by the ultraviolet rays of sunlight, resulting in various syntheses, and such syntheses result in the appearance of the smog-forming substance or substances. Sulfur dioxide as the primary irritant gas has been fairly well ruled out of consideration for many reasons, among which is the fact that it is said not to be a major air component. (On days of good visibility the maximal concentration of sulfur dioxide is 0.05 p.p.m., and on days of reduced visibility this value may rise to 0.3 p.p.m.) (Los Angeles County Pollution Control District, 1951.) According to present opinion, the oxides of nitrogen and hydrocarbons (mostly olefins), both arising from combustion processes, particularly of petroleum products, are involved in the significant photochemical changes that take place in the air (Cadle & Magill, 1951; Faith, 1954; Haagen-Smit & Bradley, 1953a,b; Los Angeles County Pollution Control District, 1954). Under laboratory conditions this reaction between oxides of nitrogen and hydrocarbons in the presence of ultraviolet light produces materials which can cause the same eye-irritating effects on humans and damaging effects on plants as those observed under natural daylight conditions during smog. For example, exposing non-irritant Los Angeles night air, which contains these precursor chemicals, to artificial ultraviolet light produces the same kind of irritant material (Littman, Ford & Endow, 1956). All these chemical reactions are associated with the production of appreciable increases of ozone in the air. There is some question, however, as to whether ozone itself is the noxious agent. In fact, the specific cause of eye irritation in Los Angeles is not known although many substances have been suspected. Furthermore, it is altogether possible that a single substance is not responsible for all three phenomena—eye irritation, vegetation damage and air haze. It is equally possible that if ill health is produced by Los Angeles smog, this may be due to air pollutants other than those that cause the eye irritation, vegetation damage and haze.

The high frequency of smog days has aroused the Los Angeles populace. The people are deeply concerned over (*a*) the possibility that some day a level of air pollution may be reached capable of causing severe illness among susceptible individuals; (*b*) the possibility that long exposure to the local air pollutants has an adverse health effect which may not appear until after many years; (*c*) the annoyance and worry caused by the eye irritation. It is interesting in connexion with the last cause of concern to speculate about the development of symptoms and of disease due to stress itself and the resulting adrenal-cortical overactivity (Foulger, 1952; Selye, 1957).

Poza Rica, 1950

Poza Rica is an oil refining and natural gas processing town in Mexico, near the coast of the Gulf of Mexico, with a population (in 1950) of about 22 000. On 24 November 1950 it was seriously affected by an accidental spillage of hydrogen sulfide gas, which caused the illness of 320 persons and 22 deaths (McCabe, 1951; McCabe & Clayton, 1952). The episode started with the accidental mishandling of natural gas processing machinery that resulted in the spillage and spread of a large amount of hydrogen sulfide gas over the residential area of the community. The situation was aggravated by a weather inversion associated with slight lateral air movement.

Within 10-20 minutes after the spillage began, the gas reached the people, causing them to become severely ill. The source of the gas was stopped 20-25 minutes after it first appeared. It was during this very short period of time that the sicknesses began and deaths occurred. The effects of the gas were those characteristic of hydrogen sulfide inhalation (Ahlborg, 1951; Mitchell & Davenport, 1924). The symptoms included loss of the sense of smell, cough, dyspnoea, conjunctival irritation, nausea and vomiting, severe headache and vertigo. Four survivors had sequelae of nervous origin, two developing neuritis of the acoustic nerve. Of the other two, one developed dysarthria due to a neurological effect on his tongue, and the other had marked aggravation of pre-existing epilepsy. The poison gas affected all ages and both sexes. No further analysis of the age or sex distribution is possible from the published report of the incident (McCabe & Clayton, 1952).

In the one autopsy that was performed the findings were non-specific, typical of death due to acute poisoning from any of a number of very noxious gases. There was congestion and haemorrhage of the lungs with slight oedema. Punctate haemorrhages were found in the brain.

Haff disease

During the four years 1924-27, inclusive, outbreaks of a peculiar malady ascribed to air pollution occurred among fishermen along the east coast of Prussia facing the Baltic Sea. There were 600 such cases in 1924, 176 in 1925, 16 in 1926, and 19 cases in 1927. The condition was termed " Haff disease " after the names of the communities in which the cases occurred —Frischer Haff, Kursicher Haff and Pomerischer Haff[1]. The disease occurred among fishermen who worked on the low-lying land along the lagoons. It also affected the dogs and birds that fed on the same land. The disease was manifested by muscle cramps, haematuria and anaemia, and thus resembled arsine gas poisoning (Lintz et al., cited by Batta et al., 1933, page 183; Pinto et al., 1950; Steel & Feltham, 1950). No air samples were collected for study to confirm the presence of arsine. A possible source of the gas was from the action of sewage-contaminated sea-water upon the arsenic-containing iron pyrites present in processed material that was discarded from a nearby factory.

Discussion

As a result of reading the foregoing accounts of air pollution incidents the reader can have no doubt that the ambient air of thickly settled and highly industrialized communities, and even of less crowded places, can occasionally be contaminated by large quantities of foreign substances that may be noxious, and that such a situation can cause epidemics of non-fatal and fatal sickness. In all places in which such incidents occurred and were adequately documented retrospective investigation generally revealed that similar situations had occurred previously. The earlier incidents, however, either lacked adequate documentation or were not recognized as crises at the time. This fact impels one to speculate about how many such episodes have occurred in places other than those for which adequate documentation exists and in how many communities they may be taking place even now. It is reasonable, however, to assume that other air pollution epidemics have occurred and not been recorded. The very nature of the condition, by virtue of its gradual development, as well as the usual manner of keeping incidence records of sickness and death, makes this probable, for an air pollution episode that is associated with adverse effects upon health is generally not recognized until it terminates—if it is recognized at all.

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[1] The word *Haff* is German, signifying a long, shallow lagoon separated from the open sea by a narrow sand bar or barrier beach.

Comparative study of the few incidents for which there are suitable records leads to the conclusion that a single pattern does not fit them all.

TABLE 9

COMPARISON AMONG THREE MAJOR AIR POLLUTION CRISES, MEUSE RIVER VALLEY, 1930, DONORA, PENNSYLVANIA, 1948, LONDON, 1952

	Meuse Valley, 1930	Donora, 1948	London, 1952
Weather	Anticyclonic, inversion, and fog	Anticyclonic, inversion, and fog	Anticyclonic, inversion, and fog
Topography	River valley	River valley	**River plain**
Most probable source of pollutants	Industry (including steel and zinc plants)	Industry (including steel and zinc plants)	**Household coal-burning**
Nature of the illnesses	Chemical irritation of exposed membranous surfaces	Chemical irritation of exposed membranous surfaces	Chemical irritation of exposed membranous surfaces
Deaths among those with pre-existing cardio-respiratory disease	Yes	Yes	Yes
Time of deaths	Began after second day of episode	Began after second day of episode	**Began on first day of episode**
Ratio of illnesses to deaths	Not available	75 : 1 to 300 : 1	Illness rates not in expected proportion to that of deaths
Autopsy findings	Inflammatory lesions in lungs included parenchyma	**Inflammatory lesions in lungs did not include parenchyma**	Inflammatory lesions in lungs included parenchyma
Suspected proximate cause of irritation	Sulfur oxides with particulates	Sulfur oxides with particulates	Sulfur oxides with particulates

There are, however, some striking similarities among three of the episodes for which we have documentation—namely, the Meuse Valley, Donora and London incidents (see Table 9). (We shall omit the Poza Rica incident, Haff disease and the Los Angeles smog because their patterns are essentially different from those of the others.)

The Meuse Valley and Donora episodes were much alike, while the London episode, though following a basically similar pattern including the same weather conditions, was different from the other two in several interesting ways. First, in regard to topography, we found that whereas the Meuse Valley communities and the city of Donora are located in deep river valleys, London is situated on a gently sloping plain. Secondly, we found that in both the Meuse Valley and Donora the largest single source of air pollution was heavy industry, including steel plants and zinc reduction plants, whereas in London industrial sources were of lesser significance and the use of coal in the individual households was the major source. Thirdly, in regard to the illnesses themselves, there were some similarities and some differences. In all three episodes the induced illnesses involved chemical irritation of the eyes, nose, throat and respiratory tract, with a consequent effect upon the heart in susceptible persons as a result of the respiratory tract irritation. There were two important differences between the health effects in London as compared to those in the Meuse Valley and Donora. These were (*a*) the time during the episode that deaths occurred, and (*b*) the ratio of the number who became ill to the number who died. In the Meuse Valley and Donora outbreaks, deaths did not begin until after the pollution had been collecting for two days. In the London episode, however, the excess deaths began on the first day, and even within the first 12 hours. This observed difference possibly may have been due to, among other reasons, a more rapid piling up of air pollutants in London than in the other two places, or to a high degree of susceptibility to severe reactions in the population of London as compared with the population of the other two places. In regard to the ratio of the number of persons who became ill to the number who died, it is to be noted that if one considers the *total* number of persons who were ill during the episode in Donora, the proportion of illness frequency to deaths becomes about 300 to one. If only the *very severely ill* are considered the proportion turns out to be about 75 to one. For the Meuse Valley incident the data do not permit an estimation of a corresponding ratio, since numerical data on sickness frequency were not reported, although the number of deaths was known. Competent authorities on the London episode of 1952 noted that morbidity rates did not increase in a proportion which one might have expected with such a known mortality (England and Wales, Ministry of Health, 1954).

Some important differences were noted in the autopsy reports. In the very few autopsies that were done in Donora it was found that the respiratory tract changes reached down only as far as the deeper bronchi and the bronchioles. In both the London and Meuse Valley autopsies the findings included inflammatory lesions throughout the tracheobronchial tree and the lung parenchyma.

It appears, therefore, that the Meuse Valley and Donora episodes were similar to each other in many ways, and differed in some significant respects from the London episode. Furthermore, whereas the first two events were associated with specific factory air effluents, it is emphasized that the London episode was characterized by pollutants arising from household coal-burning. The latter pollutants can be considered in the present state of our knowledge as chemically non-specific materials that act as irritants. So characteristic is this of urban centres throughout the world that it is referred to in one report as " typically urban " and " relatively featureless. " This is not to imply that the cause in London eventually will not be found to be a specific agent (or group of agents). It is suggested, rather, that in the London episode the air pollutants, when considered in terms of their human effects, closely resembled those present in the air of many large urban areas. These pollutants were quite similar to those of many other urban areas in that they are irritating to the exposed living membranes (of the eyes, nose, throat and respiratory tract). For example, although Los Angeles air pollution is chemically different from that of London (Lodge, 1957) the two resemble each other in their effects on man since each causes irritation of exposed living membranes. The acceptance of the concept of a " relative unity " of physiological response (Henderson & Haggard, 1943) to chemically different air pollutants, especially as such pollutants occur in urban industrialized centres, will probably serve to simplify the study of human health effects of air pollution. Such a concept will permit us to consider the quality and quantity of the pollutants in terms of their irritant effects, i.e., " units of irritancy. "

It is true, as pointed out later in this chapter, that in order to take economically reasonable steps for the control of air pollution for health reasons it is necessary that the specific chemical substances producing symptoms and illness should eventually be known. Steps leading to the acquisition of this information may be taken later, after we have studied the illnesses caused or aggravated by irritant air pollution.

Mechanism of Action of Air Pollutants

The effects of air pollution on human health generally occur as a result of contact between the pollutants and the body. The major exceptions to this generalization include the absorption of solar radiation by pollutants, causing a decrease of the intensity of these rays that may reach man, and the emanation of ionizing radiation from some radioactive isotopes with the adverse action of these ionizing rays on the body. These and other exceptions are discussed more fully later in this review.

Normally, bodily contact occurs at the surfaces of the skin and exposed membranes. Except in the unusual instances of very strongly irritant vapours and of some agents that are strongly allergic to the skin (e.g., *Rhus toxicodendron*), the skin as a point of contact with air pollutants is of much less physiological significance than are the membranous surfaces of the body that are in contact with the ambient air. Such contact with exposed membranous surfaces can be of utmost importance both because these surfaces are particularly sensitive to injury and because of their high absorptive capacity compared to that of the skin. In discussing air pollution effects upon the membranes primary consideration, it seems to us, must be given to the irritant qualities of these substances. Such airborne gases, vapours, fumes, mists and dusts may have their bodily contact with, and may cause irritation of, the membranes of the eyes, nose, nasal accessory sinuses, throat, larynx, tracheobronchial tree and lung parenchyma. Some irritants even reach the mucosa of the digestive tract.

Irritation manifests itself through the operation of the body's protective mechanisms. In the case of the eye surfaces, the conjunctiva and cornea, the earliest reaction is discomfort due to a sense of burning. Blinking and excessive lachrymation then commence and cleanse the eye of the offensive material quite effectively. The irregular air channels of the nose direct a large portion of the air-borne pollutants so that they impinge against mucosal surfaces. These moist surfaces together with the vibrissae remove part of the pollutants, preventing them from reaching deeper into the respiratory tract. The normal mucoid secretions present on the walls of the nasal cavities and the throat and the increment of such secretions induced by the irritant serve to protect the epithelium. The filtering action of the nose, in addition to catching a very small portion of inhaled gases, can be expected to hold back the larger solid and liquid particles, especially those that measure 5μ or more in size (Drinker & Hatch, 1954 ; Eisenbud, 1952).

Since the sense of smell is a nasal function and provides a modicum of protection against air pollution effects, it is mentioned at this point even though it is discussed in detail later in this review. It is, generally speaking, a poor protection against air pollution health effects. For example, the sense of smell serves only fairly well as a warning mechanism in the case of sulfur dioxide gas, an air pollutant present in many urban atmospheres (Amdur et al., 1953). Carbon monoxide, since it has no odour, is another gaseous air pollutant against which the sense of smell fails to protect.

Irritant air pollutants that reach the larynx in high concentration may cause the glottis to close abruptly and prevent further passage of air, a warning to the recipient to remove himself to a less irritant atmosphere.

A significant protective action takes place on the surface membranes of the tracheobronchial tree through the production of mucus and through the action of the cilia. The whip-like action of the cilia pushes upwards and out towards the throat those particles and dissolved gases that are trapped in the mucus (Cralley, 1942; Dalhamn, 1956; Dalhamn & Rhodin, 1956). When they reach the throat they are expectorated or swallowed. The cough mechanism also acts to remove undesirable material from the tracheobronchial tree and from the deeper parts of the lung. When a highly irritant gas reaches the membranes of the trachea or bronchi, it will paralyse the cilia and even cause desquamation of the superficial layers of the epithelial lining (Dalham, 1956; Dalham & Rhodin, 1956). There is evidence that repeated irritation of this membrane can lead to hyperplastic and metaplastic changes of the cells (Auerbach et al., 1956; Kotin, 1957; Niskanen, 1949). The relation of the metaplastic changes to the genesis of bronchial cancer is not known at the present time.

Irritant air pollutants

The part of the respiratory tract upon which an irritant gas acts is dependent in part upon the water solubility of the gas (Flury & Zernik, 1931; Henderson & Haggard, 1943). An inhaled irritant gas that is highly soluble in water is likely to be dissolved in the aqueous fluids in the upper portion of the respiratory tract and to exert its effect in that part of the airway where, as it happens, the epithelium is more resistant to injury than deeper in the respiratory tract. However, such solution of the gas can be greatly modified by the presence of aerosols in the inhaled air. Thus, a gas which by itself would not normally be carried beyond the trachea and large bronchi may be absorbed by or adsorbed on aerosols and, if those aerosols are of small enough diameter, conveyed in that manner into the deeper parts of the lungs, even as far as the alveolar walls (Dautrebande, 1951a; Gordieyeff, 1955; La Belle, Long & Christofano, 1955). Upon reaching the deeper parts of the airways the gas-laden aerosols may impinge upon the walls to exert their action, or desorption may occur in the airways themselves, allowing the gas in free molecular form to act upon the epithelial lining. There are many unexplained and unexplored factors involved in these and closely related phenomena. Two such factors readily come to mind. An irritant gas that is absorbed by the mucosa of the upper part of the respiratory tract may lead to reflex spasm of the distant bronchioles, producing severe symptoms. Secondly, very fine dust particles of some materials that *per se* are generally considered to be " inert " in terms of physiological or pharmacological action have been shown to be capable of causing bronchoconstriction, apparently by virtue of an unexplained local irritative action (Dautrebande, 1951a).

When more than one potentially irritant gas or aerosol is present in the inhaled air, the resulting irritant effects vary. The result of the combination of several irritants may be merely the addition of the effects that each would produce by itself or the resulting effect may be greater (synergistic) (Amdur, 1954) or less (antagonistic) (Stokinger, 1956) than the mere sum of the effects each irritant would produce alone. The mechanism that accounts for such variation in action is not entirely clear at this time.

The severity of tissue response to an irritant gas is not always dependent upon the product of the concentration of the gas and the duration of exposure. Thus, exposures to very low concentrations of irritant gases may be continued for long periods of time and cause practically no observable effect, whereas the same total amount of irritant gas if inhaled in a single breath can cause a great deal of tissue damage (Henderson & Haggard, 1943). This concept points up the importance of taking into account the peak levels of irritant gas concentrations when considering air pollution effects upon the lungs. In fact, since momentary high levels of concentration may have great physiological significance, it has been suggested that there is no relationship between daily " average " figures for air concentration levels of irritant gases and the observed irritative lung effects.

Experimental studies of the effects of irritant gases upon the lungs of man and animals have shown that these effects can be severe. Although all the irritant gases that may be present as pollutants in the ambient air are not known, some gases have been singled out as probable offenders. For example, in some past acute air pollution episodes sulfur dioxide gas was seriously considered to be the culprit. We shall discuss only two prototype gases, sulfur dioxide and ozone, for they are gases that have received a great deal of attention as air pollution irritants. Further investigation will establish whether or not these two gases are, in fact, major offenders.

Sulfur dioxide

Sulfur dioxide appears in the air of most urban communities, its major source being the combustion of sulfur-bearing fossil fuels. This gas is likely to be present in appreciable amounts in the air of communities in which coal, especially of the so-called lower grades, and some fuel oils are the principal sources of heat and power. This is in contrast to those places where natural gas and very low sulfur-bearing fuel oils and their products constitute the main fuel. For instance, in New York City, one of the world's largest cities, it has been estimated that 1 500 000 short tons[1] of sulfur dioxide are discharged yearly from the burning of the equivalent of 32 000 000 short tons of coal (Greenburg & Jacobs, 1956). The varying sulfur contents of fuels used in the USA are shown in Tables 10-13. When

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[1] 1 short ton = 0.9 metric ton.

TABLE 10

SULFUR CONTENT IN WEIGHT PERCENT OF COAL MINED IN THE USA, 1952 AND 1955

Kind of coal	1952			1955		
	Minimum	Average	Maximum	Minimum	Average	Maximum
Bituminous	0.2	1.8	8.7	0.3	2.0	7.9
Anthracite [a]	0.2	0.6	1.1	0.4	0.6	1.4
Lignite [b]	0.4	1.0	2.8	No data	No data	No data

Source: Aresco & Haller (1953); Aresco, Haller & Abernethy (1956).

[a] Based on figures available for Pennsylvania only.

[b] Based on all coal mined in North Dakota not, however, specified as lignite in the reference sources.

TABLE 11

SULFUR CONTENT BY WEIGHT PERCENT OF MOTOR GASOLINE IN THE USA, WINTER 1956-57

Grade of gasoline	Sulfur content by weight percent		
	Minimum	Average	Maximum
Regular-price, octane number, research O.N. 89.1	0.000	0.063	0.428
Premium-price, octane number, research O.N. 96.7	0.000	0.051	0.207

Source: Blade (1957b).

TABLE 12

SULFUR CONTENT BY WEIGHT PERCENT OF BURNER FUEL OIL USED IN THE USA, BY GRADE OF OIL, 1956

Burner oil grade	Sulfur content by weight percent			No. of samples analysed
	Minimum	Average	Maximum	
1	0.01	0.149	0.96	152
2	0.05	0.381	1.4	164
4	0.23	0.801	2.2	37
5	0.42	1.448	4.7	60
6	0.29	1.563	5.25	138

Source: Blade (1956).

TABLE 13

SULFUR CONTENT BY WEIGHT PERCENT IN DIESEL FUEL USED IN THE USA, BY GRADE OF FUEL, 1956

Diesel fuel grade	Sulfur content by weight percent			No. of samples analysed
	Minimum	Average	Maximum	
1-D	0.008	0.168	0.78	256
2-D	0.06	0.363	1.06	216
4-D	0.12	0.346	1.00	13

Source: Blade (1957a).

sulfur with a molecular weight of 32 combines with oxygen to form sulfur dioxide the resultant material has a molecular weight of 64, double that of the original elemental sulfur. Thus, in calculating the amount of sulfur dioxide that is potentially derived from the burning of sulfur-bearing fuel, the proportion of the original weight of the fuel that is present as sulfur must be multiplied by a factor of two to obtain a truer concept of how much sulfur dioxide may be discharged.

It has been shown that at a rather low level of concentration, a level not infrequently found in air, inhalation of sulfur dioxide in both experimental animals and man results in temporary spasm of the smooth muscle of the bronchioles; somewhat higher concentrations cause increased mucus production on the walls of the upper airways; still higher concentrations result in severe inflammatory responses in the mucosa, with desquamation of the surface epithelium. These irritant effects of sulfur dioxide, especially bronchiolar spasm, are aggravated by cold air. The reactions to these effects adversely modify lung function (Amdur,1954; Amdur et al., 1953; Greenwald, 1954; Lawther, 1955; Pattle & Cullumbine, 1956).

A moderate degree of resistance may develop from continued exposure to this gas. Such resistance is found among industrial workers who are continually or repeatedly exposed to the gas in their occupations (Anderson, 1950). This effect is believed to be due to the outpouring of increased amounts of mucus on to the walls of the tracheobronchial tree, serving to protect the epithelium (Henderson & Haggard, 1943). According to a report of one experiment, animals did not appear to develop such immunity after exposure to sulfur dioxide (Weedon, Hartzell & Setterstrom, 1939). It is not entirely clear under what conditions we may expect a reduced level of reaction to sulfur dioxide in man and under what other conditions the opposite—hyperirritability and hypersensitivity—may occur. Hypersensitivity and hyperirritability that are sometimes found in cases of exposure to sulfur dioxide gas are discussed below.

Under certain conditions, related partly to the availability of solar energy and partly to the coexistence in the air of some potentially catalytic metallic compounds, some of the air-borne sulfur dioxide gas is oxidized to become sulfur trioxide, an extremely irritant mist rather than a gas. A portion or each of these two gases in the presence of water vapour (or water) form sulfurous and sulfuric acids, respectively. It is likely that some of the air-borne sulfur trioxide comes directly from the same combustion process that gives rise to the dioxide. In some urban areas fog has been shown to be associated with the presence of the trioxide in appreciable amounts in relation to the quantity of sulfur dioxide that is present (Ellis, 1931). Inhalation experiments with the trioxide in both animals and man show that it is a very strong irritant, much stronger than the dioxide, causing severe bronchospasm at relatively low levels of concentration. Furthermore, there is a marked degree of synergism in the physiological reaction that becomes manifest from exposure to low levels of both gases at the same time (Amdur, 1957).

Ozone

Ozone is a gas that has an irritant action in the respiratory tract, reaching much deeper into the lungs than the sulfur oxides. The origins of ozone that is found in the air have not been clarified, but it is likely that combustion and sunlight are involved in its production.

Studies on experimental animals have shown that ozone inhalations, at levels of concentration significantly below those inducing fatal pulmonary oedema, can result in the later manifestation of resistance to ozone irritation. In the animals previously exposed to the low levels of ozone, subsequent administration of doses that ordinarily would be lethal do not result in death. The exact mechanism of this response is not at present known. Even more important, it has been discovered in our laboratories that animals subjected for a period of months to daily inhalations of sublethal doses of ozone develop fibrotic thickening of the walls of their bronchioles. Similar observations have been reported for sulfur dioxide inhalation. Although the full significance of this finding for man is as yet unknown, it is noted that there is a striking similarity between these histological observations and some of the findings in early stages of chronic bronchitis in man (Reid, 1956).

Non-irritant pollutants

Non-irritant noxious air pollutants generally have their effect on the body after they are absorbed from the lungs and are deposited elsewhere in the body. It is obvious that the speed with which this absorption occurs will be influenced by the nature of the inhaled material, among other factors. It is, perhaps, less obvious that the degree of absorption of a non-irritant pollutant can be modified by such adventitious factors as the co-existence in the inhaled air of other pollutants that may be irritant. One interesting report recently suggested that lung irritation due to a noxious agent enhances the absorption rate into the blood of other potentially noxious agents (Taplin et al., 1951). In the case of air-borne carcinogens, which may exert their action locally in the respiratory tract, the irritant quality of a second inhalant may, again, be of great significance. In this instance the irritant may be strong enough to cause partial paralysis of ciliary movements on the tracheobronchial wall, and in this manner permit an increased amount of time for contact to continue between the potential carcinogen and the susceptible epithelium (Kotin, Falk & Tremer, 1957). It may be of further significance that denudation of the surface layers of epithelial cells caused by an irritant may expose the cancer-susceptible deep-lying cells to more intimate contact with the carcinogenic agent (Kotin, 1957).

Non-irritant gases that reach the alveoli of the lungs are absorbed into the blood in accordance with well-known physical and physiological principles. The rate of absorption depends upon many factors, the more significant of which are the following: (*a*) the air concentration of the gas; (*b*) the solubility of the gas in the tissue fluids and blood; (*c*) the amount of the gas already present in the blood and tissues; and (*d*) the duration of exposure to the gas (Flury & Zernik, 1931; Henderson & Haggard, 1943). From the lungs the gas is carried by the blood to other parts of the body, where it may act upon susceptible tissues. A portion of the absorbed gas that is carried in the circulating blood is excreted by the lungs. Another portion combines with other substances in the body, changes its form, and may be excreted through the kidneys.

Of the solid and liquid materials that are discharged into the air from all sources, a large part generally consists of particles whose size and mass are of such a nature that it settles out of the air very rapidly. This action leaves behind the much smaller particles that settle out more slowly, dependent in part upon air movement. These latter liquid and solid particles (aerosols), since they are air-borne for an appreciable period of time, may be inhaled. Of the aerosols that are inhaled, generally only those of the size of about 1 μ and less may reach the alveoli of the lungs and, of these, maximal alveolar retention takes place for particles whose diameters measure between 1.0 μ and 0.6 μ and those that measure below 0.2 μ (Brown et al., 1950; Landahl, 1950; Van Wijk & Patterson, 1940). Retention of solid and liquid particulate materials depends in part on the rate and depth of respiration (Brown, 1931; Landahl, 1950; Landahl, Tracewell & Lassen, 1953; Wilson & La Mer, 1948) and similarly upon the relative density of the material being inhaled (Davies, 1949, 1952, 1955). The solid aerosols are absorbed into the blood from the alveoli mainly in proportion to their solubility in the surface and tissue fluids of the lung parenchyma.

It has been suggested that they may perhaps be modified by those fluids and thus become more readily soluble. Some solid aerosols, and possibly some oily mists, remain virtually insoluble in the aqueous liquids that line the alveolar walls and are engulfed by tissue-wandering cells which carry them into the lymphatic stream. From that stream they are deposited in pulmonary lymphatic depot points or in the lympn glands (Fenn, 1921). Some insoluble particles pass through the alveolar walls into the lymph channels without having been phagocytized by the wandering white cells (Drinker & Field, 1933). Solid particulate materials, such as silica, asbestos and some forms of carbon, when present in appropriate quantities, may, however, be capable of exerting a noxious (fibrotic) local action in the interstitial areas of the lung and in the lymphatic tissues.

Dissolved aerosols that reach the circulating blood are carried in this manner to other vital organs. Some of this material is excreted, mostly by the kidneys and the intestinal tract. Some accumulates in various specific organs and reaches a level of concentration that may exert a toxic action. Other potentially noxious agents are neutralized through the body's protective mechanisms.

Except for the unusual effect of beryllium and its compounds, of radioactive materials, and possibly of manganese compounds, systemic toxic effects among humans have not been reported from long-continued exposure to low levels of community air pollution. Furthermore, with the exceptions of the incident at Poza Rica and of Haff disease, described above, the reports of *acute* air pollution episodes indicate that the damaging effect upon health was due to a local action on exposed membranes and, when the respiratory tract was affected in some susceptible persons, this effect upon the lungs led to a secondary adverse action upon the heart. From this finding, however, many believe that it is dangerous to assume that systemic effects from community air pollutants do not occur except for the three unusual situations (beryllium, radioactive isotopes, and manganese). Unsuspected systemic changes may, perhaps, be revealed by newer and more refined methods of investigation only now being explored.

Even before a search is begun for any systemic health effects due to air pollutants, however, research workers are looking for evidence of early change in the lungs. Since experimental animals and human volunteers have demonstrated measurable changes of lung function due to the inhalation of relatively low concentrations of certain irritant gases, and because of some of the clinical observations of lung irritation found in acute episodes of air pollution, plans are now being made to study the lung function of people who are exposed to relatively low levels of irritant *community* air pollutants. For this purpose the precise prodecures of the pulmonary physiologist are being modified for testing people in their communities (Comroe, 1950). In these investigations due consideration will be given to all other known factors that may give rise to functional changes of the lungs.

In these studies search will be made only for physiological changes. Even if physiological abnormalities are found, their clinical significance will still be open to question. The margin of safety present in the normal respiratory system may be so large that although functional impairment occurs it may not be severe enough to cause symptoms of disease. It is reasonable to expect, however, that should such relatively minor impairments of lung function be found, the extra burden may be functionally intolerable for some people—e.g., those persons already burdened by malfunction of the lungs or heart.

The inadequacy of most techniques for the field observation of slight changes in lung function is magnified in the study of other organs. Preliminary studies of the eye irritation phenomenon on Los Angeles, done with currently available techniques, have yielded no objective evidence of injury. It seems that the protective mechanism provided by the tear glands limits the irritation. Experimental work has indicated that the air-borne gaseous irritants that cause eye irritation probably have their action intensified by the simultaneous presence of aerosols almost always found in ambient air (Dautrebande, 1950, 1951b, 1952).

In recent months attempts have been made in Los Angeles to correlate both the occurrence and severity of human eye irritation with certain measurements of air pollutants. The many unmeasured psychological factors inherent in a procedure involving the subjective symptom of Los Angeles eye irritation make the use of this technique of limited value (Renzetti & Gobran, 1957). An attempt is now in progress to measure these psychological factors themselves. Furthermore, because of the many unsolved problems involved in the subjective nature of the reply obtained from a person as to whether or not his eyes are irritated at a given moment of time, a search is currently being made for more objective tests. One of the most promising avenues being explored for this purpose is the use of a dissected ciliary nerve preparation made from the eye and adnexa of a suitable animal. In this preparation, air-borne irritative chemical stimulation is applied to the eye surface and the resultant nerve impulse is measured electronically. If this procedure turns out to be a satisfactory one for the low concentrations of the gaseous air pollutants that are known to be present in the Los Angeles ambient air, then it will be possible to correlate those observations with the human subjective response. This procedure will attempt to provide a more objective standard.

Further study of the systemic effects of air pollution is still in its early stages. The studies that are being done are based partly on the concept that both the concentration and the time of exposure are significant factors in any kind of *systemic* poisoning that may result from inhaled substances. From occupational disease studies it is known that there is generally a maximal amount that one may allow the worker to breathe with impunity (*A.M.A. Arch. industr. Hlth,* 1956). As has been outlined previously, these data, originating from studies of industrial exposures, cannot be safely applied to community air pollution. There has been only a single report, thus far, listing recommended safe limits of exposure for potentially toxic pollutants in community air (Riaznov, 1954).

In order to clarify the subject of the systemic health effects of community air pollution, the following types of studies are being conducted: (*a*) community health studies; (*b*) studies of groups of individuals for whom health records are available (having been collected for other purposes); and (*c*) laboratory investigations. In the community health studies, efforts are being directed towards the determination of the health patterns of communities, particularly where the source of the pollutants can be easily traced, as, for example, to a single industrial plant. Data on sickness occurrence and sickness aggravation, as well as mortality data, will be measured against air pollution levels to determine if there are significant correlations. Great care must be used in such studies to ensure that all possible etiological factors, other than air pollution, are also considered and appropriate corrections made of the data. Although correlations are being made between health data and levels of air concentration of specific pollutants (and groups of pollutants), we do not know with certainty that we are using the correct air pollution information for this purpose. For this reason we are exploring the feasibility of also using a series of broadly based indexes of air pollution levels. In developing these indexes for a community, the following complex factors are included, all of which contribute to the level of air pollution that may be reached: type and size of industry, fuel consumption, weather, topography and population density.

War veterans on disability pensions for chronic disease of the heart or lungs, members of health insurance programmes and other groups for whom continuing health records are available are being studied for air pollution health effects. A serious problem in such investigations is created by the mobility of population, which makes difficult, but not impossible, the use of the place of residence of an individual as an indicator of exposure to air pollutants. Previous places of residence must also be taken into account, for we do not now know the length of time of exposure to low grade air pollution that may be significant in terms of health and disease. It is even possible that such exposure at certain times of life—e.g., infancy—may be critically related to disease in later life.

Although other organ systems of the body are not being neglected in these investigations of community health, primary emphasis is again being directed towards the component parts of the respiratory tract because, as noted above, significant irritation of exposed mucous membranes are very apt to occur in that system. It is also suspected that the heart may show significant changes as a secondary effect of lung irritation. Because of the tremendous reserve power present in both the heart and the lungs, however, we suspect that primary disease of those organs is less likely to be found in these studies than, possibly, evidence of aggravation of disease in persons whose hearts or lungs are already diseased.

The laboratory investigations that are being made to elucidate the systemic as well as the local irritative effects of air pollutants are aimed primarily at providing us with better techniques for discovering physiological changes in people. A second purpose of these studies is to provide us with information about mechanisms of action of the noxious agents.

One set of laboratory investigations is devoted to the study of the *in vitro* effect of single air pollutants and groups of pollutants upon body enzyme systems. A second is devoted to studying the effects of such noxious agents upon tissue cultures and involves the observation of cell growth and multiplication, and cell metabolism. Other laboratories are engaged in studying the effects upon suitable intact laboratory animals of known and suspected air pollutants, including ozone and other oxidants, sulfur oxides, organic sulfides, phenolic compounds and nitro-olefins. Some detailed laboratory investigations are being done on the evaluation of the carcinogenic potential of community air pollutants. Since the work of all these laboratories has been in progress for a short time, it is too early to discuss their results.

Systemic effects

Returning now to a consideration of the systemic health effects of air pollution, we find that among the agents that may cause such effects and are found as pollutants are the following: beryllium and its compounds, manganese compounds, carbon monoxide, fluorides, radioactive isotopes, aeroallergens, carcinogens and insecticides.

Beryllium

Shortly after the appearance of the first published reports of the occurrence of the specific chronic lung disease (chronic pulmonary granulomatosis) in persons who had been exposed to beryllium and its compounds in their work even many years before (Hardy & Tabershaw, 1946), it was found that a few people living in the communities near the industrial plant developed the same disease (Chesner, 1950; Eisenbud et al., 1949; Goldwater, 1951) even though neither they nor any member of their households had a connexion with the plants. Although there were records of air pollution complaints of offensive odours and of *local* irritative effects on exposed membranous surfaces (such as of the eyes, nose, throat, and tracheobronchial tree) due to factory air discharges, to our knowledge this was the first instance in the USA in which a characteristically *specific* systemic disease ascribable to air pollutants from industry was found among people not directly concerned with the factory operations. Among the sources of the beryllium compounds that contaminated the community air, even though in minute quantities, were factories that were either extracting the metal from the ore or those that were handling compounds of beryllium used in the manufacture of fluorescent electric light lamps.

Manganese

Prior to the first reports of the beryllium lung disease cases, there was a Norwegian report that pneumonia occurred with an unusually high frequency among people living near a manganese processing plant (Elstad, 1939). In that report it was suggested that the increase of case frequency was due to the inhalation of manganese compounds present in the ambient air as a result of the factory air discharges. In a recent report from Italy, there was suggestive confirmation of such a medical relationship between air-borne manganese compounds and pneumonia (Pancheri, 1955). It is difficult to be sure of the causal significance of manganese in this situation since the pneumonitis in these cases is very similar clinically to cases not associated with manganese as an etiological factor. Such a difficulty does not arise in the case of the substance beryllium in relation to the disease for which it is responsible since the chronic beryllium lung disease is quite distinctive as a granulomatous lung condition.

Carbon monoxide

Carbon monoxide, an odourless and colourless gas that has its major origin in the incomplete combustion of carbonaceous materials, has long been known as a noxious inhalant that has its effects because of a strong affinity for combining with the haemoglobin of the blood. Thus, it appears that carbon monoxide is noxious only by virtue of this combination that leaves less haemoglobin available to fulfil its normal function, the carrying of oxygen. When a sufficient amount of carbon monoxide attaches itself to the haemoglobin in the circulating red blood cells, the effect is that there is then less haemoglobin available to combine with oxygen, and this results in the reduction of the amount of oxygen available to the tissues. Although the danger from carbon monoxide inhalation under certain conditions, as in some occupations, has been recognized for some time, opinion is still divided as to the noxious significance of this gas in community air (Bloomfield & Ishell, 1928; *Bull. N. Y. Acad. Med.*, 1926, 1931; Cambier & Marcy, 1928; Connolly, Martinek & Aeberly, 1928; Drinker, 1938; Florentin, 1927; Gunn & Hitschfield, 1951; Ives et al., 1936; Kling, 1938; Regan, 1932; Sayers & Davenport, 1937; Sievers et al., 1942; Silverman, 1929; Stern, 1945; Wilson et al., 1926). This question has become even more important in recent years because of the ever-increasing use of the automobile, which discharges appreciable amounts of the gas in the engine exhaust (Portheine, 1954). It is doubtful that the concentrations of carbon monoxide in city streets often reach or exceed 100 p.p.m., the amount generally considered as the industrial upper limit of safety when exposure may continue for an eight-hour period (*A.M.A. Arch. industr. Hlth*, 1956; Flury & Zernik, 1931; Henderson & Haggard, 1943). It is possible, however, that the levels of carbon monoxide that are reached in the streets may affect some especially susceptible persons, such as those already suffering from a disease associated with a decrease of oxygen-carrying capacity of the blood (e.g., anaemia), or those suffering from cardio-respiratory disease. The extra burden that is placed on the body by the reduction of the oxygen-carrying capacity of the blood induced by carbon monoxide may cause injury to vital organs in such persons. People who are already burdened by the presence in their blood of variable amounts of carbon monoxide because of tobacco smoking or possibly because of exposure in their occupations, may also be adversely affected by the extra amount of the gas that they may be required to inhale with the carbon monoxide-contaminated air. This is a subject that requires further exploration.

By its very nature as a relatively non-cumulative poison and its mode of action in the organism (Henderson & Haggard, 1943), it would seem that carbon monoxide can cause acute poisoning as a result of exposure to high air concentrations of the gas, but not chronic poisoning due to long-continued exposure to relatively low concentrations. The possible exception to this statement may be found in the instances described above of persons who are already affected by decreased oxygen-carrying capacity of the blood or by cardio-respiratory disease. Recently, however, the controversy has reopened as to whether or not this gas can cause disease from continued (or intermittent) exposure to the low concentrations present in the ambient air (Drinker, 1938; Grut, 1949; Lewey & Drabkin, 1944; Noro, 1945; Sumari, 1949; Symanski, 1942). Relatively strong evidence for the affirmative has come from Europe. In the USA, however, the more common belief is that " chronic carbon monoxide poisoning " does not occur.

Mention of the potential health hazard from carbon monoxide as a community air pollutant, and the fact that the automobile is one of its major sources, bring to mind the potential lead poisoning hazard from the combustion of the compound tetraethyl of lead that is present in most gasoline to the extent of about 3 ml per gallon. [1] Studies of the air contamination from lead from this source indicate that the amount present in the community air is minimal and probably has no clinical significance, especially when it is considered in the light of the amount of lead man obtains from food and water (Cholak, Schafer & Hoffer, 1950, 1952; England and Wales, Ministry of Health, 1930; Kehoe, personal communication, 1947; Kehoe, Thamann & Cholak, 1934, 1936; *Publ. Hlth Bull. (Wash.)*, 1926; Sayers et al., 1924; Valentine, 1936). It is possible that an opposite conclusion may be reached from continued study of the problem in some selected places in the world.

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[1] US gallon = 3.8 l.

In a recent study of this subject the following interesting observations were reported: (*a*) in ordinary city driving the automobile exhaust contains only from 20 % to 60 % of the lead that it burns as the tetraethyl compound; (*b*) the lead appears in the exhaust as PbCl.Br. and as complexes of that compound with ammonium chloride; (*c*) the particle size of the discharged lead compound varies from those of less than 0.01 μ to those of many millimetres, with about 5 % by weight accounting for the smallest particles; (*d*) at high speed, such as that which occurs on highways, the lead compounds in the exhaust gases are greater than the amount burned, the excess arising from the walls of the engine and the exhaust piping where it had accumulated during the period of slower city driving (Hirschler et al., 1957).

The automobile engine is responsible for other air pollutants in addition to carbon monoxide and compounds of lead. In the section of this report devoted to a description of Los Angeles air pollution, it is noted that it is currently believed that the automobile is the major source of the precursors of smog. That is to say that the automobile exhaust supplies the hydrocarbons and oxides of nitrogen to the air, and that these materials when acted upon by solar energy result in the production of materials that bring about eye irritation, vegetation damage and air haze. That the automobile engine also produces chemical substances which are known to be carcinogenic to experimental animals, and the significance of this fact to man, are discussed in a later section of this chapter.

The subject of automobile engine exhaust gases and aerosols in relation to the entire air pollution problem is one that requires much more intensive study. Beginnings in this direction have been made in regard to the components of these exhausts, showing that the composition varies with many factors, including engine design, engine operation, engine maintenance and type of fuel used (Faith, 1954; Fitton, 1954). Plans are now being made to intensify long-range studies of the effects of these materials upon experimental animals.

Fluorides

Fluoride compounds comprise another group of specific substances that are known to be present in the air of some communities and that may be significant to human health. The fluorides known to reach the ambient air range from those which are relatively soluble in body fluids to those which are virtually insoluble, and from the extremely irritant and corrosive hydrogen fluoride to relatively non-reactive compounds. Discharges of fluorides into the air have their major sources in the following types of industrial processes: artificial fertilizer manufacture, aluminium production and some forms of steel production.

It is clear that those fluorides which are chemically highly reactive will be irritating to the exposed surfaces of the body (skin and some membranes) when they are present in the air in sufficient concentration. Exposures to such highly reactive fluorides are extremely rare. Less, however, is known about the effects on human beings of those inhaled fluorides that are not surface irritants and whose action, if there is any, depends upon their absorption by the blood from the lungs to act elsewhere in the body. The action of minute amounts of fluoride salts that are ingested in food and water, and possibly inhaled, during the period of life when the teeth are developing, is not germane to the problem of air pollution. It is sufficient to note that such teeth are less prone to decay. The significance, however, of the effect of the fluorides upon bones and joints at all other times in the life of man is still a matter of controversy (Agate et al., 1949; Green, 1946; Greenwood, 1940; Kilborn, Outerbridge & Lei, 1950; Largent, 1950; Roholm, 1937). The fact that adverse health effects due to fluorides have been shown to occur in animals that graze in the vicinity of factories discharging fluorides into the air does not signify that a similar action can be expected in people living in the same neighbourhood. Among the cogent reasons for this is that the grazing animal probably obtains the greater portion of fluoride from eating fluoride-contaminated vegetation and from drinking grossly fluoride-contaminated water. It has been shown, for example, that whereas forage originating in areas in which the air is known not to be contaminated with fluorides may contain as little as from 2 p.p.m. to 75 p.p.m. of the fluoride residual, expressed as fluorine, similar forage, grown in areas where fluoride is known to exist as an air pollutant, often has as much as 500-1000 p.p.m. of the fluoride (National Academy of Sciences, 1956).

Radioactive isotopes

This discussion of radioactivity, since it applies only to the problem of air pollution, is limited to a consideration of radioactive materials that may be present in the air as gases or aerosols which emit such ionizing radiations as alpha and beta particles, gamma rays, neutrons and other high energy quanta. From the point of view of current use, possibility of adventitious formation, and quantities available, the most important radioactive isotopes that may conceivably reach the ambient air, and are generally available in compounds rather than as elements, include iodine-131, phosphorus-32, cobalt-60, strontium-90, carbon-14, sulfur-35, calcium-45, gold-198, radium-226 and uranium (US National Bureau of Standards, 1953).

Although it is true that man has been exposed to ionizing radiations from radioactive isotopes and other sources since the beginning of his history, only with the discovery of the existence of natural radioactive isotopes and of their production by man, and especially with the development of the atomic reactor, have these radiation sources become of real importance by nature of their sudden increase in amount. In view of the world's need for new sources of energy, and for other obvious reasons, the isotopic sources of radioactivity are likely to continue to increase in amount. The potential sources of radioactive air pollutants are: (*a*) the increased removal of soil and rock overburden that lies over many natural underground radioactive deposits; (*b*) the settling out from the upper reaches of the atmosphere of radioactive aerosols that had their origin in the detonation of radioactive weapons (radioactive fall-out); (*c*) medical and scientific use of radioactive isotopes; (*d*) agricultural and industrial use of radioactive isotopes as tracers; (*e*) research and commercial power reactors; and (*f*) experimental accelerators. Exposure to radiation from materials arising from these sources, with or without exposure to radiation for clinical purposes, can cause tissue and organ injury unless suitable precautions are taken. It is emphasized that the biological effects from such radiations, regardless of their sources, are cumulative.

The possible radiation effects of these noxious materials, and the degree of delay in their manifestation, are varied and depend upon such factors as the amount reaching the body, their chemical and physical nature, their locus of initial contact with the body, their radioactive half-lives, the kind of radioactivity that they emit, the level of energy of the radioactivity that they emit, and, finally their movement and metabolism within the body and their excretion rates (Anderson et al., 1947; Bloom, 1948; Bugher, 1957; Cowan, Farabee & Lowe, 1952; Hamilton, 1947, 1948; Hunter & Ballou, 1951; Hyde, 1906; Marinelli, Quimby & Hine, 1948; Morgan, 1949; Skipper, Nolan & Simpson, 1951; Voegtlin & Hodge, 1949). Among the more important health effects described as being caused by radioactive isotopes are the following: blood abnormalities, including anaemia, leukaemia, leukopenia and haemorrhagic diseases (Browning, 1949; Cronkite et al., 1955; Cronkite, Bond & Durham, 1956; De Bernardi & Lendini, 1953; Fletcher, 1954; Henn & Leibetseder, 1955; Jacobson & Marks, 1947; Jacobson, Marks & Lorenz, 1949; Looney et al., 1955; Lorenz & Congdon, 1954; Lynch, 1951; Moshman, 1951; Peller & Pick, 1952; Snell & Neel, 1949; Tsuzuki, 1955); skin changes, including erythema, atrophy, ulceration, pigment changes, temporary and permanent alopecia and cancer (Cronkite et al., 1956; Ellinger, 1951; Knowlton et al., 1949; Moritz & Henriques, 1952); bone changes—bone necrosis and bone sarcoma (Andrews, 1955); thyroid changes—lowered thyroid activity (Warren, 1956); gonad changes—genetic effects and varying grades of sterility (Warren, 1956); embryo effects—congenital malformations (Warren, 1956); and lung changes—fibrosis and cancer (Andrews, 1955; Davies, 1952; Eisenbud & Harley, 1953, 1955; Warren, 1956).

The recognition of the health hazards from radioactive isotopes that may contaminate the ambient air has led to the institution of controls at the possible sources. These control measures have apparently been quite successful, with the rare exceptions of some scattered incidents that occurred in a few remote locations (Arnold, 1954; Cronkite et al., 1956; Kimura, 1954; Lapp, 1954, 1955 a, b, c, 1956; Ravina, 1954; Tsuzuki, 1955; Yamasaki & Kakehi, 1954). It appears, therefore, that adverse health effects from this kind of air pollutant have been kept to a minimum. The only important subject in which opinion on this point is divided is that of the possible genetic effect (Muller, 1955).

Allergenic agents

There is general recognition by the health professions that the air we breathe is the natural carrier of living and dead solid and liquid microscopic organic materials which may act as allergens. The human tissue reactions to such allergens occur predominantly in the skin and the respiratory tract. When the reaction takes place in the respiratory tract, the mucosa of the nasal cavities and the involuntary muscle tissue of the bronchioles are particularly affected (Cooke, 1947). These organic allergenic materials have their origins in living things, be they plants, yeasts, or moulds, or in animal emanations such as danders, hair, fur, or feathers (Bernton, 1923; Bernton & Thorn, 1933; Brown, 1936; Cadham, 1924; Cooke, 1947; Feinberg & Little, 1935; Figley & Elrod, 1928; Hopkins, Benham & Kestin, 1930). Certain industrial operations may add to this allergenic community air load; such operations include baking, cotton milling, fur processing, hair processing, carpet making, felt hat making, feather processing, flour milling, jute processing, leather processing, rope making, castor bean processing, shoe making, soap making, woodworking and tobacco processing (Dublin & Vane, 1942).

Although industrial operations contribute to the problem of air-borne allergens, the size of that contribution is relatively small when considered in relation to the problem as a whole. Air-borne allergens are primarily of natural origin. That is to say, allergic disease resulting from aero-allergenic materials is mostly not man-made and, therefore, is controlled with much more difficulty than if it were created by man. As would be expected the chemical nature of these naturally produced noxious materials is very complex, consisting of compounds of very high molecular weight. In some of the man-made situations that cause allergic reactions or possibly conditions simulating allergic reactions, the chemical nature of the agents involved turns out to be relatively simple (Morris, 1956).

Sulfur dioxide gas has been described as inducing such an allergic reaction (Cooke, 1934; Dowling, 1937; Romanoff, 1939). What is surprising, however, is the paucity of reports of such complaints in the light of the large number of persons potentially or actually exposed to this gas in their normal living. Probably what occurs more frequently with exposure to sulfur dioxide is a hyperirritability of the bronchial mucosa, developing after earlier exposures to the gas, which results in an exaggeration of the normal reaction to relatively low doses of the gas. (It is noted elsewhere in this review that repeated inhalation of this gas may also lead to an apparent resistance to its irritant action.) There is also the possibility that the micro-organisms usually present in the respiratory tract will find that the gas-injured mucosa serves as an excellent nidus for an increased rate of reproduction, and that this overproduction leads not only to severe infections but also to an allergic response to the absorbed proteins derived from the micro-organism.

Using animals (Amdur & Mead, 1955) and human volunteers, experiments have shown that inhalation of low concentrations of sulfur dioxide results in a spasmodic tightening of the bronchioles that generally goes unrecognized in non-experimental situations. This is generally a simple reflex reaction to an irritant gas. It is possible that this latter mechanism was the immediately precipitating factor in the fatalities that occurred among bronchial asthmatic and cardiac patients in both the Meuse Valley and Donora episodes. In those cases such bronchiolar spasm could have resulted in an aggravation of an existing serious defect in respiratory gas exchange. The significance to humans of the bronchiolar constriction, when it is not associated with existing cardio-respiratory disease, is not clear. It probably results in decreased efficiency of lung function. However, the normal cardio-respiratory reserve is so great that the effect of the spasm is generally not noted by the subject. Even the medical observer will not note that it exists unless special techniques are used to elicit the fact. This is a subject which needs careful study with suitable instrumentation.

Beryllium, in the form of its compounds, is a substance that is said to cause its peculiar pulmonary granulomatous reaction (Hardy & Tabershaw, 1946; Machle, Beyer & Gregorius, 1948) as the result of an allergic response to its inhalation (Eisenbud, Berghout & Steadman, 1948; Eisenbud et al., 1949; Sterner & Eisenbud, 1951). Finely divided metallic cobalt, by virtue of the fact that its inhalation in man is associated with a lung disease that is roentgenographically similar to that due to beryllium, is also considered as a possible cause of the same type of allergic manifestation (Miller et al., 1953). We know of no case reports of cobalt lung disease occurring among persons outside of the industrial establishments themselves.

The commercial fur dye, paraphenylenediamine, is an allergenic agent capable of causing dermatoses in addition to bronchial asthma (Heimann, 1942; Mayer, 1929; Mayer & Foerster, 1929; Rich, 1924; Silverberg & Heimann, 1941). In view of the manner in which this dye is commercially used, the possibility of its appearing in the ambient air outside of the fur-processing institutions is a real one, although we have never seen reports of such cases.

Factories processing the castor bean for the extraction of the oil discharge a powdery material into the air that is a strong allergen. This material has been the cause of bronchial asthma in people living near the factories (Bernton, 1923; Figley & Elrod, 1928).

More important than these unusual situations in the study of air pollution is the strong probability that air pollutants ordinarily considered as irritants for exposed membranes may, under some special circumstances, act as the stimulating agents for the allergic response in susceptible persons (Cooke, 1934; Romanoff, 1939). This concept may also include the phenomenon wherein the chemically highly reactive air pollutant combines with body proteins, and the newly formed complex then acts as a true allergen (Burrage et al., 1956; Landsteiner, 1945). An interesting case in point is the syndrome termed " Yokohama asthma, " a clinical entity that recently occurred in and about Yokohama, Japan. In this instance a number of people living in the area developed bronchial asthmatic attacks when they were exposed to the industrially polluted air of some Japanese cities, including Yokohama. They had no attacks when they were removed from the communities in question (Huber et al., 1954).

Carcinogenic agents

Exposures to specific occupational situations have long been known to induce human cancer in susceptible individuals (Ask-Upmark, 1955; Baetjer, 1950; Brockbank, 1941; Cruickshank & Squire, 1950; Flinn, 1931; Gehrmann, 1954; Gross & Alwens, 1940; Heller, 1950; Hoffmann, 1931; Hueper, 1942, 1954; Machle & Gregorius, 1948; Mancuso, 1951; Peller, 1939; Pott, 1775; Shapiro et al., 1953; Snegireff & Lombard, 1951; Spitz, Maguigan & Dobriner, 1950; Warren, 1942), and it is reasonable to expect that the specific causative chemical and physical agents are present outside the respective work-places, in the community ambient air, and in this manner affect persons other than the workers concerned. Of course, the amounts of the culpable agents that escape from the work-place are small (Bourne & Rushin, 1950; Hueper, 1957a, b; Mancuso & Hueper, 1951). However, even though the amounts of known potential carcinogens found in the outdoor air may be very small, the significance to man of even these amounts may be considerable, since the human lifetime exposures are of a high order of magnitude (Blacklock et al., 1954).

But the carcinogenic substances that come from industrial sources are not the only agents responsible for the cancer problem associated with air pollution. For instance, the polycyclic organic compound, 3,4-benzpyrene, well known as a potent carcinogen for experimental animals (Hartwell, 1951), and considered as a standard by which carcinogenicity of other chemical agents is measured, has been found in the air of many communities (Cooper, 1953; Cooper & Lindsay, 1953; Dikun, Shabud & Norkin, 1956; Falk & Steiner, 1952; Sterner, 1954; Waller, 1952). In addition to this agent, other similar organic compounds of varying carcinogenic potency have been found in the air (Kotin, Falk & Thomas, 1955).

It has been reported that this group of compounds, and particularly 3, 4-benzpyrene, is present in urban air in appreciably larger quantities than in rural air (Stocks & Campbell, 1955; Waller, 1950, 1952). These compounds have their origin in the incomplete combustion of hydrocarbons and other carbonaceous materials, a common occurrence since such combustion very often does not reach completion—i.e., form carbon dioxide and water—unless special efforts are exerted toward this end. In fact there was recently reported the successful experimental production of 3, 4-benzpyrene, as well as related polycyclic organic compounds, from the incomplete burning under suitable conditions of even simple, short, straight-chain hydrocarbons (Tebbens, Thomas & Mukai, 1956a,b). Such polycyclic materials have been shown also to be present in biologically significant quantities in exhaust discharges that come from the internal combustion engine, both Diesel and gasoline, when operating under suitable conditions (Commins, Waller & Lawther, 1956; Fitton, 1956; Kotin et al., 1954; Kotin, Falk & Thomas, 1954, 1955; Mills & Seipp, 1955).

Fortunately for man, these carcinogens are not highly stable, being destroyed at variable rates by other air constituents and by sunlight. Their potential for harm, however, cannot be denied, for the duration of time of their integrity in the ambient air is appreciable, long enough to be of possible significance (Kotin, Falk & Thomas, 1954). It has even been noted that some of the non-carcinogenic compounds in the air may act as adjuvant agents for the carcinogens by carrying the latter to the target tissue (Kotin, 1957). Note, however, must be taken of the strong variation in species' susceptibility to carcinogens, even among experimental animals. As Hartwell (1951) states: "It is, therefore, dangerous to attempt to carry over without reservation, to man, conclusions based on animal experiments."

In addition to the air-contained polycyclic organic compounds that are known to be carcinogenic when appropriately supplied to suitable species of experimental animals, it is reported that the air may carry certain aliphatic hydrocarbons that also are carcinogenic (Fieser et al., 1955; Hendry, Horner & Walpole, 1951; Kotin & Falk, 1956; Kotin, Falk & Thomas, 1956).

In the animal experiments in which the various carcinogenic chemicals are used, the target tissue that responds with a malignant cancer growth may be in the respiratory tract or it may be at another site. In connexion with the fact that respiratory tract cancer has been experimentally produced by these materials, and the strong current belief that these materials are discharged into the air in larger amounts in recent years as a result of urbaniration and industrialization, it is noteworthy that recent epidemiological zeports have shown that human lung cancer frequency has been steadily increasing over many areas of the world, especially in urbanized industrialized communities (Clemmesen & Busk, 1947; Doll, 1953; Dorn, 1953; Kreyberg, 1954; Phillips, 1954). The possible causal relationship of tobacco smoking to this increase is receiving world-wide attention, as the voluminous literature on the subject attests (Breslow et al., 1954; Clemmesen, 1954; Clemmesen, Nielsen & Jensen, 1953; Cornfield, 1951; Cutler & Loveland, 1954; Doll & Hill, 1952, 1954; Gsell, 1951; Hammond, 1954; Hammond & Horn, 1954; McConnell, Gordon & Jones, 1952; Mueller, 1939; Nielsen & Clemmesen, 1954; Oberling, 1954; Ochsner et al., 1952; Sadowsky, Gilliam & Cornfield, 1953; Schrek et al., 1950; Wynder, 1952; Wynder & Graham, 1950). The subject of tobacco smoking and its manifold possible health effects, although involving a problem of " personal " air pollution, falls outside the province of our immediate consideration, except in so far as the smoke so produced adds to the pollution of the air breathed by bystanders. Of some importance in connexion with tobacco smoking is the recent report suggesting that if cigarette smoking does, in fact, contribute to the increased frequency of human lung cancer, it cannot account for all of that increase (Eastcott, 1956; Hyde, 1906); urban air pollution, it is argued, also contributes to the frequency of the disease (Hueper, 1957b; Stocks & Campbell, 1955). Thus it would appear that in human cancer, as in other disease, we often deal with conditions that have multiple causation, such multiplicity being operative both when the disease is considered as a mass human phenomenon and when it occurs in an individual.

Currently in progress is a study of the carcinogenic potential of particulate air-borne material collected from some urban areas. These urban centres are known to have widely varying rates of lung cancer incidence. By testing the carcinogenic potential of the air pollutants, it is expected that a link will be added to the chain of evidence of the possible causal relationship between air pollution and human lung cancer.

The respiratory tract (and specifically the lung) is not the only body area that may show cancer development as a possible result of specific air pollutants. The skin may be a target through direct contact with the potential carcinogen (Brockbank, 1941; Cruickshank & Squire, 1950; Pott, 1775; Schrek et al., 1950). Less well appreciated is the concept that carcinogens impinging originally on one body surface may be transported through the circulating blood and act upon a target tissue elsewhere in the body (Copp, Axelrod & Hamilton, 1947).

In this section of the review we have stressed the point that air pollution may lead to cancer in humans. There is, however, one aspect of air pollution that could militate against cancer induction. This is the modification of solar rays caused by air pollutants. The potential of the ultraviolet light from the sun to induce cancer is generally accepted by epidemiologists (Dubreuilh, 1907; Hofmann, 1931; Hueper, 1942; Hyde, 1906; Lawrence, 1928; Molesworth, 1927; Mountin, Dorn & Boone, 1939; Roffo, 1933; Shapiro et al., 1953). The presence in the ambient air of pollutants that screen out these harmful rays, thus reducing this risk from sunlight, is discussed elsewhere in the report.

Insecticides

The use of special sprays and dusts to destroy insect pests (and, to a lesser extent, the chemical agents that destroy unwanted vegetation) is an accepted practice in many countries. The insecticides are generally disseminated in the air to attack flying insects and, in some cases, the active ingredients have their effect only after they settle on fixed objects, such as vegetation, where they may come into contact with the insects. In either case the ambient air is *purposely* polluted with a material that is known to be noxious for insects and that may also be poisonous for man.

The recent introduction into agricultural practice of the potent organic phosphorus agents and the chlorinated hydrocarbon compounds has created a potentially serious air pollution problem in farming areas. Among the better known organic phosphorus insecticides are parathion, TEPP, and malathion. The chlorinated hydrocarbon group includes chlordane, DDT, and dieldrin. All these materials are toxic for man and must be used with caution. The organic phosphorus insecticides can be absorbed in toxic amounts by the skin and the eye membranes, as well as by the lungs. They cause an irreversible inhibition of the enzyme cholinesterase, and as a result large quantities of acetylcholine accumulate in the body. This accumulation of acetylcholine accounts for the majority of the symptoms observed in people exposed to these compounds (Rowher & Haller, 1950; Sumerford et al., 1953). The chlorinated hydrocarbon insecticides do not all have the same type of toxic effect in man. They can affect the central nervous system or they may attack other vital organs (US Department of Health, Education & Welfare, 1956).

When the dangers of these poisons are recognized and appropriate steps are taken for control, cases of poisoning from this intentionally induced air pollution need not occur.

Some Special Problems

There are some special problems of air pollution health effects that do not fall into the categories presented above. These include (*a*) the effects of sunlight upon air pollution and the effects of air pollution upon sunlight; (*b*) dust storms; (*c*) ingestion of air pollutants; and (*d*) odours.

Sunlight

In considering sunlight in relation to air pollution there are two matters that require evaluation: (*a*) the effect that air pollutants have on the ability of the sun's rays to reach man on the surface of the earth, and (*b*) the modifying effect that the rays exert upon the pollutants. (This review will omit consideration of the part payed by air pollution in reducing the amount of effective sunlight reaching vegetation.)

There is ample experimental evidence that sunlight is sharply reduced by air pollutants because of their screening action (Ashworth, 1931; Blum, 1952; Clelland, Cumming & Ritchie (1952); Drinker, Thompson & Choate, 1930; Hand, 1949; Ives, 1933; MacGregor, 1947; *Nature (Lond.)*, 1909; *Publ. Hlth Rep. (Wash.)*, 1930; *Safety Engng*, 1946; Tonney, Hoeft & Somers, 1930). The screening effect is especially marked for those ultraviolet rays whose wave-lengths measure between 2970 Å and 3020 Å, the rays that have special significance as antirachitic factors (Ashworth, 1931; Barrett, 1935; Blum, 1952; Drinker et al., 1930; Schrader, Coblentz & Korff, 1929). It is noted, however, that clothing also serves to prevent these rays from contacting the skin, which they must reach in order to have the effect of changing the precursor to antirachitic vitamin D. It is doubtful, therefore, that the loss of ultraviolet rays due to air pollution generally needs to be considered of moment in regard to the prevention of rickets.

The loss of ultraviolet light, however, may be of importance when its bacteriostatic and bactericidal powers are considered (Bayne-Jones & Van der Lingen, 1923; Del Mundo & McKhann, 1941; Gates, 1930; Higgins & Hyde, 1947; Miller et al., 1948; Ronge, 1948; Wells, 1955; Whisler, 1940). The significance of this in relation to the ecology of man is not known since the noxious effect of these rays would not be expected to affect all micro-organisms equally and certainly could not be expected to attack only the pathogens. Air-borne micro-organisms, as well as those lying on the surface of the body's airways, may also be directly affected by the active chemical air pollutants themselves. The effects of both sunlight and air-borne chemicals upon bacteria and viruses are open to further investigation (Fincke, 1883; Holman, 1913).

The decrease of sunlight caused by air pollution may have a significant effect upon man's well-being by adversely affecting him psychologically, causing a sense of depression in susceptible individuals (*Bull. N. Y. Acad. Med.*, 1931; Clark, 1929; Opie, 1921; Stoner, 1913; Wallin, 1913; Winslow & Herrington, 1935). The full significance of this possibility is not known at the present time.

The decreased light and the haziness of the air due to air pollutants, particularly those occurring over urban areas, has added a problem to aviation, for they result in decreased visibility and the obliteration from view of familiar landmarks. Such a loss of visibility can extend even to ground level, create an extra hazard for automobile traffic as well and lead to an increased number of accidental injuries.

It has already been noted that sunlight can chemically modify air pollutants. This is particularly significant in relation to the oxidant type of pollution, since the sun's rays may lead to the production in the ambient air of an irritant gas from a non-irritant one (Haagen-Smit & Fox, 1954). This type of photochemical change is involved in the reaction between air-borne hydrocarbons and the oxides of nitrogen, both believed to have their major source in the exhaust gases from automobiles. So very important is this matter considered in Los Angeles, for example, that serious efforts are being directed to deal with this highly specialized problem of automobile exhausts. For this purpose consideration is being given to three possible avenues of approach: (*a*) change of the motor fuel; (*b*) change of the engine; and (*c*) chemical alteration of the automobile exhaust gases after they have been formed in the engine.

Air-borne sulfur dioxide can also be oxidized, so that the dioxide is changed to sulfur trioxide, the sun providing the energy for the reaction (Johnstone, 1952). Other factors are undoubtedly concerned with this chemical change.

A peculiar phenomenon that may occur with the loss of solar energy caused by air pollutants is the intensification of the degree of pollution. This may occur by virtue of the fact that the decrease of sunlight causes intensification of the weather inversion that probably originally led to the initial accumulation of the pollutants.

Dust storms

On occasion man is forced to breathe air contaminated with large amounts of dust having its origin in the soil. This situation arises when heavy winds are combined with the presence of a very dry soil, such as occurs after a drought, to cause a dust storm. Such a dust storm occurred in the west-central portion of the USA in 1935, and the human health significance of the phenomenon was studied (Brown, Gottlieb & Laybourn, 1935). On the basis of a study of the morbidity and mortality records of the area, it was determined that acute respiratory disease frequency increased sharply during and shortly after the dust-laden air developed, and this was interpreted as having been caused by the siliceous dust that acted as a mechanical irritant to the mucosa of the respiratory tract. No unusual pathogenic organisms were found in the dust *per se* in that investigation. However, in connexion with the recent occurrence of a tornado, it was found that human skin sensitivity tests with appropriate doses of histoplasmin showed a marked rise in the frequency of " positive " reactions among those who had been in the vicinity of the storm (Manos, personal communication, 1957). This could be interpreted as signifying that the organism causative of the disease histoplasmosis was spread by the air-borne materials in the tornado to many persons who had been uninfected previously.

Studies carried out among dwellers in the Sahara Desert in North Africa, who are exposed to desert sand storms, have shown that, although the sand blown about by the wind was made up mostly of free silica (silicon dioxide), silicosis was not found among the exposed population (Policard & Collet, 1952). Post-mortem studies indicated that appreciable amounts of silica particles, however, were present in the lung. These particles were located along the arterial trunks; but nodules characteristic of silicosis were absent. One explanation offered for this finding was that the silica grains from the desert sand were " old " particles and that only freshly fractured silica particles can induce the typical silicotic pulmonary nodule (Bagnold, 1941; Briscoe, 1949; Briscoe, Matthews & Sanderson, 1936-37; Clelland et al., 1952; Dempster & Ritchie, 1952; Heffernan, 1935, 1948; Policard, 1949; Wilson & La Mer, 1948).

Ingestion of air pollutants

In the Meuse Valley, Donora, and London episodes nausea and vomiting occurred as symptoms among some of the affected individuals. Although the mechanism for the production of these symptoms could have been one of many, the specific one involved was not clarified in the reports and analyses of the episodes. It is possible that cough itself, a very common symptom among the ill persons, may have been of such severity that it induced nausea and vomiting. The reaction could have been due to a reflex response of the vomiting mechanism to an irritation of the mucosa of the tracheobronchial tree by air pollutants. It is also possible that the passive movement of secretions from the tracheobronchial tree, induced by coughing as well as by the ciliary action, may have accumulated enough of the irritant material so that significant contact occurred with the sensitive mucosa of the posterior portion of the throat, and this in turn resulted in nausea and vomiting; or that the mucus itself present in the throat caused the symptoms. Finally, particulate air pollutants, with or without adsorbed irritating gases, that are screened out in the nose and throat may have been swallowed and the irritative response of the gastric mucosa to those substances was the cause of nausea and vomiting. In connexion with the last hypothesis, it is of interest to note that animal experimental work with inhalation of the sulfur oxides has shown that gastric irritation can occur to the extent of even producing mucosal haemorrhage.

The swallowing of potentially toxic material that has its source in air pollution may take place not only from the pollutants that are arrested in the nose and throat but also from the ingestion of water or food that has been exposed to the settling of air-borne particles. Reference is made at this point to another section of this chapter, which discusses the effect of the ingestion of fluorides by cattle and in which it was noted that the animals obtained much of the fluoride by ingestion rather than through inhalation. The ingestion of noxious materials from the settling of air pollutants may be of importance particularly in regard to the extremely noxious poisons, such as some radioactive isotopes. The likelihood of simple irritant injury is not very great from ingestion of air-borne acidic matter in view of the fact that the gastric juices themselves are generally highly acid. It is likely that systemic poisoning from the gastro-intestinal tract absorption is of greater importance.

Rain and snow wash materials out of the air, some being dissolved in the aqueous medium (Blanchard, 1950; Gunn & Hitschfield, 1951; Langmuir, 1948; McCully et al., 1956). The watery solution seeps into the soil, from which it may be absorbed by growing vegetation. The ingestion of such contaminated vegetation by animals or by man may cause ill health if the material in solution is of a noxious nature. Some of the contaminated rain-water or melting snow may reach acceptable potable water supplies and create a health hazard from that source. It has been suggested that, in addition to the generally salubrious effect of rain as a scavenger of air pollutants, the pollutants may actually modify the occurrence of precipitation of moisture from the air (Gunn & Phillips, 1957).

There are some metallic elements which, although present in the body in exceedingly small quantities, may still be significant to the health of man. Food ingestion accounts for the major portion of the body content of these elements. A portion, however, may have its origin in air contamination, reaching the body through inhalation or ingestion (Chambers, Foter & Cholak, 1955; Gibson & Selvig, 1944). Some of these metallic elements are known to be involved in enzymatic reactions and other vital body functions (Kleiner, 1951). The importance of the others to the health of man is not clear at this time. Only recently have analytical methods been developed for the study of the presence of the minute quantities of these metals in tissues (Butt et al., 1954; Koch et al., 1956; Tipton, 1956; Tipton et al., 1957).

Odours

The ordinary man relies mainly on his senses of sight and smell to inform him of the presence of pollutants in the air that he breathes. Smell is especially important in this regard. However, it is unsatisfactory to depend upon smell as an indicator of the possible dangers in the air we breathe since many noxious gases and aerosols have no odours at all and the odour threshold of others may be relatively high. Of the gases and vapours whose odour thresholds are low, some may be toxic and others may be innocuous. Toxic gases and vapours do not necessarily have a disagreeable odour.

At times the ambient air is permeated with odours that are not associated with toxic gases. Man may find such odours objectionable on aesthetic grounds—an unhealthful situation in itself because this makes him uncomfortable and may induce nausea and vomiting. In such cases steps are taken to remove the offensive smells. Although there are many basic principles involved in odour removal (Turk, 1954) we shall consider only those that may be involved with health effects. Where it is possible to do so, the source of the odours should be removed by whatever suitable method is available. Another procedure is to use harmless masking odours that man considers more tolerable. It is also possible to introduce materials into the air that result in anaesthesia of the sense of smell, so that the undesirable odours are not perceived. When either of the latter procedures is instituted to mask odours that emanate from noxious agents, it can be harmful, for it removes the signal of warning supplied by the first odour (McCord & Witheridge, 1949; Weaver, 1950). When agents that are anaesthetic to the organ of smell are introduced into the air they may themselves cause reactions of a toxic nature.

Discussion

It is apparent from the foregoing that an appreciable amount of knowledge exists about the effects of community air pollution upon human health. This knowledge comes in part from direct studies of the air pollution health problem and in part from investigations done for other purposes. It is equally apparent that there are many aspects of the subject of the health effects of air pollution on which sound information is lacking. Many years undoubtedly will pass before we have the answers to all the questions involved.

Man-made air pollution could be entirely eliminated, but the price that civilization would be required to pay for this would be exorbitant by any standards, whether monetary or otherwise. It is unreasonable to contemplate that we could put a stop to all combustion, the chief source of man-made air pollution. It is logical, however, to consider that the clarification of the air on a qualitatively and quantitatively *selective* basis is feasible and, in some cases, highly desirable. This can be done, for example, by selectively arresting the contaminants at their source.

If we should contemplate the clarification of the air on a selective basis in the interests of safeguarding human health, then it must first be clearly demonstrated that there are specific air pollutants (or specific mixtures of air pollutants) that can cause or are causing ill health. We shall also need to know the sources of these culpable pollutants. When we are in possession of these two sets of facts, we shall be in a position to balance the importance of the health effects against the economic and other costs required to achieve air clarification. Again it is emphasized that the air that one breathes need not be in a pristine state for the purpose of maintaining good health. Not only is the kind of adulterant present significant, but the amount present is of even greater importance. We must have knowledge of the levels of concentration of air contaminants that we may breathe with safety. It will undoubtedly require a long time before this necessary information becomes available in a scientifically acceptable form.

In the meanwhile one comprehensive report on this subject has appeared. In this report there are presented the maximum allowable concentrations for community ambient air of a series of potential air pollutants. However, since the data are not based primarily on the possible health effects of the pollutants in question but rather upon economic and aesthetic effects, the figures quoted do not serve our immediate purpose (Riazonov, 1954).

As we await further information, some attempts have been made to cope with the air pollution problem, using the few data that exist. In some of the localities in which there were acute crises of air pollution, measures were instituted to prevent recurrences. The authorities in London and Los Angeles are still studying how to deal with their own particular situations.

Even larger is the problem of assessing air pollution health effects in localities in which acute episodes have not been recorded. In such localities, contaminated with relatively low concentrations of air pollutants, the health effects are very difficult to assess. Investigations have been carried out in such communities but, for many reasons, great doubt has been cast on the validity of the conclusions reached in some of those studies (Ascher, 1907a, b; Haythorn & Meller, 1938; Heimann et al., 1951; Klotz & White, 1914; Leonard, McVerry & Crowley, 1940; Mills, 1943, 1945, 1946a, b, c; Mills & Mills-Porter, 1948; Schnurer, 1938). Recent studies of the effect upon health of long-continued exposure to low concentrations of air pollutants have shown that the frequency and severity of chronic lung disease, specifically chronic bronchitis, are greater in areas of greater pollution (Daly, 1954; Hewitt, 1956; Pemberton & Goldberg, 1954). In spite of the controversy about the value of the reports of the studies, steps have been taken in many places to reduce the pollution of the air. In some places, the steps were taken, however, for economic and aesthetic, rather than for health, reasons. It is to be noted that the reduction of the amount of visible air pollutants alone—the method used when air is cleaned for aesthetic reasons—may not always be a good procedure, since it may also selectively remove some of the air-borne agents that serve to neutralize the potentially harmful agents. Thus, the procedure may result in more danger to health than that which existed before the control process was instituted. It has been suggested, for example, that the reduction of the sulfur oxide content of Los Angeles air, accomplished by the local oil refineries, contributed to a faster accumulation of eye-irritating oxidants in the community air.

REFERENCES

Abercombie, G. F. (1953) *Lancet*, **1**, 234

Abrams, H. K. (1949) *Calif. Hlth*, **7**, 1

Agate, J. N., Bell, G. H., Boddie, C. E., Bowler, R. G., Buckell, M., Cheeseman, E. A., Douglas, T. G. J., Druett, H. A., Garrad, J., Hunter, D., Perry, K. M. A. & Richardson, J. D. (1949) *Industrial fluorosis; a report*, London (Medical Research Council Memorandum No. 22)

Ahlborg, G. (1951) *A.M.A. Arch. industr. Hyg.* **3**, 247

Alexander, J. (1931) *Science*, **73**, 96

A.M.A. Arch. industr. Hlth, 1956, **14**, 186

Amdur, M. O. (1954) *Publ. Hlth Rep. (Wash.)*, **69**, 503

Amdur, M. O. (1957) *Amer. industr. Hyg. Ass. Quart.*, **18**, 149

Amdur, M. O. & Mead, J. (1955). In: *Proceedings of the third national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 150

Amdur, M. O., Melvin, W. W. & Drinker, P. (1953) *Lancet*, **2**, 758

Amdur, M. O., Silverman, L. & Drinker, P. (1952) *A.M.A. Arch. industr. Hyg.*, **6**, 305

American Association for the Advancement of Science (1942) *Fluorine and dental health*, Washington

Anderson, A. (1950) *Brit. J. industr. Med.*, **7**, 82

Anderson, E. C., Libby, W. F., Weinhouse, S., Reid, A. F., Kirshenbaum, A. D. & Gross, E. V. (1947) *Physics Rev.*, **72**, 931

Andrews, H. L. (1955) *Science*, **122**, 453

Aresco, S. J. & Haller, C. P. (1953) *Analyses of tipple and delivered samples of coal (collected during the fiscal year 1952)*, Washington (United States Bureau of Mines Report of Investigations 4972)

Aresco, S. J., Haller, C. P. & Abernethy, R. F. (1956) *Analyses of tipple and delivered samples of coal (collected during the fiscal year 1955)*, Washington (United States Bureau of Mines Report of Investigations 5270)

Arnold, J. R. (1954) *Bull. atom. Scient.*, **10**, 347

Ascher, L. (1907a), *Engng News*, **58**, 434

Ascher, L. (1907b) *J. roy. sanit. Inst.*, **28**, 88

Ashe, W. F. (1952) In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 456

Ashworth, J. R. (1931) *Investigation of atmospheric pollution ; report on observations in the year ended March 31, 1930* (Great Britain, Department of Scientific and Industrial Research), London

Ask-Upmark, E. (1955) *Dis. Chest*, **27**, 427

Auerbach, O., Petrick, T. G., Stout, A. P., Statsinger, A. L. & Muehsam, G. E. (1956) *Cancer (Philad.)*, **9**, 76

Baetjer, A. M. (1950) *Arch. Industr. Hyg.*, **2**, 487

Baetjer, A. M. & Smith, R. (1956) *Amer. J. Physiol.*, **186**, 39

Bagnold, R. A. (1941) *The physics of blown sand and desert dunes*, London

Barrett, H. J. (1935) *J. industr. Hyg.*, **17**, 199

Batta, G., Firket, J. & Leclerc, E. (1933) *Les problèmes de pollution de l'atmosphère*, Paris

Bayne-Jones, S. & Van der Lingen, J. S. (1923) *Bull. Johns Hopk. Hosp.*, **34**, 11

Bernton, H. S. (1923) *Amer. J. med. Sci.*, **165**, 196

Bernton, H. S. & Thorn, C. (1933) *J. Allergy*, **4**, 114

Blacklock, J. W. S., Kennaway, E. L., Lewis, G. M. & Urquhart, M. E. (1954) *Brit. J. Cancer*, **8**, 40

Blade, O. C. (1956) *National survey of burner fuel oils, 1956*, Washington (United States Bureau of Mines Information Circular 7762)

Blade, O. C. (1957a) *National annual Diesel-fuel survey, 1956*, Washington (United States Bureau of Mines Information Circular 7776)

Blade, O. C. (1957b) *National motor gasoline survey, winter 1956-1957*, Washington (United States Bureau of Mines Information Circular 7796)

Blanchard, D. (1950) *Trans. Amer. Geophys. Un.*, **31**, 836

Bloom, W. (1948) *Histopathology of irradiation from external and internal sources*, New York

Bloomfield, J. J. & Isbell, H. S. (1928) *Publ. Hlth Rep.*, **43**, 750

Blum, H. F. (1952) *Effect of loss of sunlight on human health.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 62

Bourne, H. G. & Rushin, W. R. (1950) *Industr. Med. Surg.*, **19**, 568

Breslow, L., Hoaglin, L., Rasmussen, G. & Abrams, H. K. (1954) *Amer. J. publ. Hlth*, **44**, 171

Briscoe, H. V. A. (1949) *Chem. Trade J.*, **124**, 478

Briscoe, H. V. A., Matthews, J. W. & Sanderson, P. M. (1936-37) *Trans. Inst. Min. Metallurg. (Lond.)*, **46**, 243

Brit. med. J., 1931, **1**, 277

Brit. med. J., 1953, **1**, 50

Brockbank, E. M. (1941) *Brit. med. J.*, **1**, 622

Brodie, F. J. (1891) *Quart. J. roy. meteorol. Soc.*, **18**, 41

Brodie, F. J. (1905) *Quart. J. roy. meteorol. Soc.*, **51**, 15

Brown, C. E. (1931) *J. industr. Hyg.*, **13**, 293

Brown, E. G., Gottlieb, S. & Laybourn, R. L. (1935) *Publ. Hlth Rep. (Wash.)*, **50**, 1369

Brown, G. T. (1936) *J. Allergy*, **7**, 455

Brown, J. H., Cook, K. M., Ney, F. G. & Hatch, T. (1950) *Amer. J. publ. Hlth*, **40**, 450

Browning, E. (1949) *Brit. med. J.*, **1**, 428

Bugher, J. C. (1957) *Amer. J. publ. Hlth*, **47**, 682

Bull. N. Y. Acad. Med., 1926, **2**, 402

Bull. N. Y. Acad. Med., 1931, **7**, 751

Burrage, W. S., Greene, J. E., Burgin, L. B., Itkin, I. H., Novey, H. S., Engler, J. & Irwin, J. W. (1956) *New Engl. J. Med.*, **255**, 79

Butt, E. M., Nusbaum, R. E., Gilmour, B. A. & DiDio, S. L. (1954) *Amer. J. clin. Path.*, **24**, 385

Cadham, F. T. (1924) *J. Amer. med. Ass.*, **83**, 27

Cadle, R. D. & Magill, P. L. (1951) *A.M.A. Arch. industr. Hyg.*, **4**, 74

California State Department of Public Health (1955) *Clean air for California*, San Francisco

California State Department of Public Health (1956) *Clean air for California; second report*, San Francisco

Cambier, R. & Marcy, F. (1928) *C. R. Acad. Sci. (Paris)*, **186**, 918

Chambers, L. A., Foter, M. J. & Cholak, J. (1955) In: *Proceedings of the third national air pollution symposium*, Los Angeles (Stanford Research Institute)

Chem. Engng News, 1948, **264**, 3692

Chesner, C. (1950) *Ann. intern. Med.*, **32**, 1028

Cholak, J., Schafer, L. J. & Hoffer, R. F. (1950) *Arch. industr. Hyg.*, **2**, 443

Cholak, J., Schafer, L. J. & Hoffer, R. F. (1952) *A.M.A. Arch. industr. Hyg.* **6** 314

Clark, R. V. (1927) *Med. Offr*, **38**, 93

Clark, R. V. (1929) *J. roy. sanit. Inst.*, **50**, 394

Clelland, D. W., Cumming, W. M. & Ritchie, P. D. (1952) *J. appl. Chem.*, **2**, 31

Clemmesen, J. (1954) *Dan. med. Bull.*, **1**, 37

Clemmesen, J. & Busk, T. (1947) *Brit. J. Cancer*, **1**, 253

Clemmesen, J., Nielsen, A. & Jensen, E. (1953) *Brit. J. Cancer*, **7**, 1

Commins, B. T., Waller, R. E. & Lawther, P. J. (1956) *Brit. med. J.*, **2**, 753

Comroe, J. H. (1950) *Meth. med. Res.*, **2**, 74

Connolly, J. I., Martinek, N. J. & Aeberly, J. J. (1928) *Amer. J. publ. Hlth*, **18**, 1375

Cooke, R. A. (1934) *J. Amer. med. Ass.*, **102**, 664

Cooke, R. A. (1947) *Allergy in theory and practice*, Philadelphia

Cooper, R. L. (1953) *Chem. & Ind.*, p. 1364

Cooper, R. L. & Lindsay, A. J. (1953) *Chem. & Ind.*, p. 1177

Copp, D. H., Axelrod, D. J. & Hamilton, J. G. (1947) *Amer. J. Roentgenol.*, **58**, 10

Cornfield, J. (1951) *J. nat. Cancer Inst.*, **11**, 1269

Cowan, F. P., Farabee, L. B. & Lowe, R. A. (1952) *Amer. J. Roentgenol.*, **67**, 805

Cralley, L. V. (1942) *J. industr. Hyg.*, **24**, 193

Cronkite, E. P., Bond, V. P., Conrad, R. A., Shulman, N. R., Farr, R. S., Cohn, S. H., Durham, C. L. & Browning, L. E. (1955) *J. Amer. med. Ass.*, **159**, 430

Cronkite, E. P., Bond, V. P. & Durham, C. L., ed. (1956) *Some effects of ionizing radiation on human beings*, Washington (United States Atomic Energy Commission)

Cruickshank, C. N. D. & Squire, J. R. (1950) *Brit. J. industr. Med.*, **7**, 1

Cutler, S. J. & Loveland, D. B. (1954) *J. nat. Cancer Inst.*, **15**, 201

Dalhamn, T. (1956) *Acta physiol. scand.*, **36**, Supplement No. 123

Dalhamn, T. & Rhodin, J. (1956) *Brit. J. industr. Med.*, **13**, 110

Daly, C. (1954) *Brit. med. J.*, **2**, 687

Dautrebande, L. (1950) *Arch. int. Pharmacodyn.*, **82**, 505

Dautrebande, L. (1951a) *L'Aérosologie*, Paris

Dautrebande, L. (1951b) *Arch. int. Pharmacodyn.*, **85**, 17

Dautrebande, L. (1952) *Physiol. Rev.*, **32**, 214

Davies, C. N. (1949) *Brit. J. industr. Med.*, **6**, 245

Davies, C. N. (1952) *Brit. J. industr. Med.*, **9**, 120

Davies, C. N. (1955) *A.M.A. Arch. industr. Hlth*, **11**, 431

De Bernardi, E. & Lendini, A. (1953) *J. Radiol. Electrol.*, **34**, 165

Del Mundo, F. & McKhann, C. F. (1941) *Amer. J. Dis. Child.*, **61**, 213

Dempster, P. B. & Ritchie, P. D. (1952) *Nature (Lond.)*, **169**, 538

Dikun, P. P., Shabud, L. M. & Norkin, V. L. (1956) *Gig. i Sanit.*, No. 1, 6

Doll, R. (1953) *Brit. med. J.*, **2**, 521, 585

Doll, R. & Hill, A. B. (1952) *Brit. med. J.*, **2**, 1271

Doll, R. & Hill, A. B. (1954) *Brit. med. J.*, **1**, 1451

Dorn, H. F. (1953) *Acta Un. int. Cancr.*, **9**, 552

Douglas, C. K. M. & Stewart, K. H. (1953) *Meteorol. Mag.*, **82**, 67

Dowling, H. F. (1937) *Med. Ann.*, **6**, 299

Drinker, C. K. (1938) *Carbon monoxide asphyxia*, New York

Drinker, C. K. & Field, M. E. (1933) *Lymphatics, lymph, and tissue fluid*, Baltimore

Drinker P. & Hatch, T. (1954) *Industrial dust*, New York

Drinker, P., Thompson, R. M. & Choate, S. P. (1930) *Trans. Amer. Soc. mech. Engrs*, **52**, 45

Dublin, L. I. & Vane, R. J. (1942) *Occupation hazards and diagnostic signs*, Washington (United States Department of Labor Bulletin 41)

Dubreuilh, W. (1907) *Ann. Derm. Syph. (Paris)*, **8**, 387

Eastcott, D. F. (1956) *Lancet*, **1**, 37

Eisenbud, M. (1952) *A.M.A. Arch. industr. Hyg.*, **6**, 214

Eisenbud, M., Berghout, C. F. & Steadman, L. T. (1948) *J. industr. Hyg. Toxicol.*, **30**, 281

Eisenbud, M. & Harley, J. H. (1953) *Science*, **117**, 141

Eisenbud, M. & Harley, J. H. (1955) *Science*, **121**, 677

Eisenbud, M., Wanta, R. C., Dustan, C., Steadman, L. T., Harris, W. B. & Wolf, B. S. (1949) *J. industr. Hyg. Toxicol.*, **31**, 282

Ellinger, F. (1951) *Ann. N. Y. Acad. Sci.*, **53**, 682

Ellis, B. A. (1931) *The investigation of atmospheric pollution ; 17th report*, London, p. 38 (Great Britain, Department of Scientific and Industrial Research)

Elstad, D. (1939) *Nord. Med.*, **3**, 2527

England and Wales, Ministry of Health (1930) *Final report of the Departmental Committee on Ethyl Petrol*, London

England and Wales, Ministry of Health (1954) *Mortality and morbidity during the London fog of December 1952*, London (Reports on Public Health and Medical Subjects, No. 95)

Faith, W. L. (1954) *Combustion and smog*, Los Angeles (Air Pollution Foundation, Report No. 2)

Falk, H. L. & Steiner, P. E. (1952) *Cancer Res.*, **12**, 30, 40

Feinberg, S. & Little, H. T. (1935) *Wisc. Med. J.*, **34**, 254

Fenn, W. O. (1921) *J. gen. Physiol.*, **3**, 575

Fieser, L. F., Greene, T. W., Bischoff, F., Lopez, G. & Rupp, J. J. (1955) *J. Amer. chem. Soc.*, **77**, 3928

Figley, K. D. & Elrod, R. H. (1928) *J. Amer. med. Ass.*, **90**, 79

Fincke (1883) *Dtsch. med. Wschr.*, **9**, 685

Firket, J. (1936) *Trans. Faraday Soc.*, **32**, 1192

Firket, M. (1931) *Bull. Acad. roy. Méd. Belg.*, **11**, 683

Fitton, A. (1954) *Pollution of the atmosphere by the exhaust gases of Diesel and petrol engines*, London (Memorandum No. 31, Fuel Research Station)

Fitton, A. (1956) *Roy. Soc. Prom. Hlth J.*, **76**, 664

Fletcher, R. D. (1949) *Weatherwise*, **2**, 56

Fletcher, W. R. (1954) *Radiography*, **20**, 19

Flinn, F. B. (1931) *J. Amer. med. Ass.*, **96**, 1763

Florentin, D. (1927) *C. R. Acad. Sci. (Paris)*, **185**, 1538

Flury, F. & Zernik, F. (1931) *Schädliche Gase*, Berlin

Foulger, J. H. (1952) *Amer. Ass. industr. Hyg. Quart.*, **13**, 70

Fry, J. (1953) *Lancet*, **1**, 235

Gates, F. L. (1930) *J. gen. Physiol.*, **14**, 31

Gehrmann, G. H. (1934) *J. Urol. (Baltimore)*, **31**, 126

Gibson, F. H. & Selvig, W. A. (1944) *Rare and uncommon chemical elements in coal*, Washington (United States Bureau of Mines Technical Paper, No. 669)

Glueckauf, E. (1951) In: American Meteorological Society, *Compendium of meteorology*, Boston, Mass., p. 6

Goldwater, L. J. (1951) *Arch. Mal. prof.*, **12**, 35

Gordieyeff, V. A. (1955) *Toxicity of vapors in the presence of aerosol carriers*, Edgewood, Md (Army Chemical Center)

Great Britain, Committee on Air Pollution (1953) *Interim report, presented to Parliament by the Minister of Housing and Local Government, the Secretary of State for Scotland and the Minister of Fuel and Power... December 1953*, London

Great Britain, Committee on Air Pollution (1955) *Report, presented to Parliament by the Minister of Housing and Local Government, the Secretary of State for Scotland and the Minister of Fuel and Power... November 1954*, London

Green, H. H. (1946) *Proc. roy. Soc. Med.*, **39**, 795

Greenburg, L. & Jacobs, M. B. (1956) *Industr. Engng Chem.*, **48**, 1517

Greenwald, I. (1954) *Arch. industr. Hyg.*, **10**, 455

Greenwood, D. A. (1940) *Physiol. Rev.*, **20**, 582

Gross, E. & Alwens, W. (1940) *Zbl. Gewerbehyg. u. Unfallverhüt.*, **27**, 1197

Grut, A. (1949) In: *Proceedings of the ninth international congress on industrial medicine*, London, p. 911

Gsell, O. (1951) *Schweiz. med. Wschr.*, **81**, 662

Gunn, R. & Hitschfeld, W. V. (1951) *Meteorology*, **8**, 7

Gunn, R. & Phillips, B. B. (1957) *J. Meteorol.*, **14**, 272

Haagen-Smit, A. J. & Bradley, C. E. (1953a) *Air Repair*, **4**, 105

Haagen-Smit, A. J. & Bradley, C. E. (1953b) *Industr. Engng Chem.*, **45**, 2086

Haagen-Smit, A. J. & Fox, M. M. (1954) *Air Repair*, **4**, 105

Haldane, J. S. (1931) *Brit. med. J.*, **1**, 366

Hama, G. M. (1950) *Industr. Hyg. Quart.*, **11**, 30

Hamilton, J. G. (1947) *Radiology*, **49**, 325

Hamilton, J. G. (1948) *Rev. mod. Phys.*, **20**, 718

Hammond, E. C. (1954) *Cancer (Philad.)*, **7**, 1100

Hammond, E. C. & Horn, D. (1954) *J. Amer. med. Ass.*, **155**, 1316

Hand, I. F. (1949) *Bull. Amer. meteorol. Soc.*, **30**, 252

Hardy, H. C. & Tabershaw, I. R. (1946) *J. industr. Hyg.*, **28**, 197

Hartwell, J. L. (1951) *Survey of compounds which have been tested for carcinogenic activity*, Washington, pp. 197-232

Haythorn, S. R. & Meller, H. B. (1938) *Amer. J. publ. Hlth*, **28**, 479

Heffernan, P. (1935) *Tubercle (Lond.)*, **16**, 397

Heffernan, P. (1948) *Tubercle (Lond.)*, **29**, 169

Heimann, H. (1942) *J. industr. Hyg.*, **24**, 322

Heimann, H., Reindollar, W. F., Brinton, H. P. & Sitgreaves, R. (1951) *A.M.A. Arch. industr. Hyg.*, **3**, 399

Heller, J. R. (1950) *Arch. industr. Hyg.*, **2**, 390

Hemeon, W. L. (1955) *A.M.A. Arch. industr. Hlth*, **11**, 397

Henderson, Y. & Haggard, H. W. (1943) *Noxious gases*, New York

Hendry, J. A., Horner, R. F., F. L. & Walpole, A. L. (1951) *Brit. J. Pharmacol.* **6**, 235

Henn, O. & Leibetseder, F. (1955) *Strahlentherapie*, **97**, 435

Hewitt, D. (1956) *Brit. J. Soc. prev. Med.*, **10**, 45

Higgins, R. A. & Hyde, G. M. (1947) *N. Y. St. J. Med.*, **47**, 707

Hirschler, D. A., Gilbert, L. F., Lamb, F. W. & Niebylski, L. M. (1957) *Industr. Engng Chem.*, **49**, 1131

Hoffmann, F. L. (1931) *The occupational incidence of cancer*, Newark, N. J.

Holman, W. L. (1913) *Amer. J. publ. Hlth*, **3**, 1210

Holman, W. L. (1914) *Mellon Institute Smoke Investigation Bulletin*, No. 9, Pittsburgh, p. 104

Hopkins, J. G., Benham, R. W. & Kestin, B. M. (1930) *J. Amer. med. Ass.*, **94**, 6

Huber, J. E., Joseph, S. W., Knoblock, E., Redfearn, P. L. & Karakawa, J. A. (1954) *Arch. industr. Hyg.*, **10**, 399

Hueper, W. C. (1942) *Occupational tumors and allied diseases*, Springfield, Ill.

Hueper, W. C. (1954) *A.M.A. Arch. Path.*, **58**, 360, 475, 645

Hueper, W. C. (1956) *Dis. Chest*, **30**, 146

Hueper, W. C. (1957a) *A. M. A. Arch. Path.*, **63**, 427

Hueper, W. C. (1957b) *Cancer (Philad.)*, **1**, 404

Hunter, H. F. & Ballou, N. E. (1951) *Nucleonics*, **9**, 62

Hyde, J. N. (1906) *Amer. J. med. Sci.*, **131**, 1

Isaac, P. C. G. (1953) *A.M.A. Arch. industr. Hyg.*, **8**, 212

Ives, J. E. (1933) *Publ. Hlth Rep. (Wash.)*, **48**, 113

Ives, J. E., Britten, R. H., Armstrong, D. W., Gill, W. A. & Goldman, F. H. (1936) *Publ. Hlth Bull. (Wash.)*, No. 224

Jacobson, L. O. & Marks, E. K. (1947) *Radiology*, **49**, 286

Jacobson, L. O., Marks, E. K. & Lorenz, E. (1949) *Radiology*, **52**, 371

J. Amer. med. Ass., 1937, **108**, 903

Johnstone, H. F. (1952) *Properties and behavior of air contaminants.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 147

Kehoe, R. A., Thamann, F. & Cholak, J. (1934) *J. industr. Hyg.*, **16**, 100

Kehoe, R. A., Thamann, F. & Cholak, J. (1936) *J. industr. Hyg.*, **18**, 42

Kennedy, H. W. (1954) *The history, legal and administrative aspects of air pollution control in the County of Los Angeles*, Los Angeles

Kilborn, L. G., Outerbridge, T. S. & Lei, Hai-Peng (1950) *Canad. med. Ass. J.*, **62**, 135

Kimura, K. (1954) *Kagaku*, vol. 24

Kleiner, I. S. (1951) *Human biochemistry*, St Louis, p. 440

Kling, A. (1938) *Rev. Hyg. Méd. prev.*, **60**, 444

Klotz, O. & White, W. C. (1914) *Mellon Institute Smoke Investigation Bulletin*, No. 9, Pittsburgh

Knowlton, N. P., Leifer, E., Hogness, J., Hempelmann, L. H., Blaney, L. F., Gill, D. C., Dakes, W. R. & Shafer, C. L. (1949) *J. Amer. med. Ass.*, **141**, 239

Koch, H. J., Smith, E. R., Shimp, N. F. & Connor, J. (1956) *Cancer (Philad.)*, **9**, 499

Kotin, P. (1957) In: *Proceedings of the air pollution research planning seminar*, Washington (Public Health Service), p. 105

Kotin, P. & Falk, H. L. (1956) *Cancer (Philad.)*, **9**, 910

Kotin, P., Falk, H. L., Mader, P. & Thomas, M. (1954) *A.M.A. Arch. industr. Hyg.*, **9**, 153

Kotin, P., Falk, H. L. & Thomas, M. (1954) *A.M.A. Arch. industr. Hyg.*, **9**, 164

Kotin, P., Falk, H. L. & Thomas, M. (1955) *A.M.A. Arch. industr. Hyg.*, **11**, 113

Kotin, P., Falk, H. L. & Thomas, M. (1956) *Cancer (Philad.)*, **9**, 905

Kotin, P., Falk, H. L. & Tremer, H. (1957) *Amer. J. Path.*, **33**, 609

Kreyberg, L. (1954) *Brit. J. Cancer*, **8**, 209

La Belle, C. W., Long, J. E. & Christofano, E. E. (1955) *A.M.A. Arch. industr. Hlth*, **11**, 297

Landahl, H. D. (1950) *Bull. math. Phys.*, **12**, 43

Landahl, H. D., Tracewell, T. N. & Lassen, W. H. (1951) *A.M.A. Arch. industr. Hyg.*, **3**, 359

Landahl, H. D., Tracewell, T. N. & Lassen, W. H. (1953) *A.M.A. Arch. industr. Hyg.*, **6**, 508

Landsteiner, K. (1945) *The specificity of serological reactions*, Cambridge, Mass.

Langmuir, I. J. (1948) *Meteorology*, **5**, 175

Lapp, R. E. (1954) *Bull. atom. Scient.*, **10**, 349

Lapp, R. E. (1955a) *Bull. atom. Scient.*, **11**, 45

Lapp, R. E. (1955b) *Bull. atom. Scient.*, **11**, 206

Lapp, R. E. (1955c) *Bull. atom. Scient.*, **11**, 339

Lapp, R. E. (1956) *Bull. atom. Scient.*, **12**, 261

Largent, E. (1950) In: *Proceedings of the first national air pollution symposium, November 1949*, Los Angeles (Stanford Research Institute), p. 129

Lawrence, H. (1928) *Med. J. Aust.*, **1**, 615

Lawther, P. J. (1955) *Lancet*, **2**, 745

Leeuwen, W. S. van (1931) *Münch. med. Wschr.*, **78**, 49

Leeuwen, W. S. van (1933) *Chem. Zbl.*, **1**, 807

Leonard, A. G., McVerry, B. P. & Crowley, D. (1940) *Sci. Proc. roy. Dublin Soc.*, **22**, 257

Lewey, F. H. & Drabkin, D. L. (1944) *Amer. J. med. Sci.*, **208**, 502

Littman, F. E., Ford, H. W. & Endow, N. (1956) *Industr. Engng Chem.*, **48**, 1492

Lodge, J. P. (1957) In: *Proceedings of the air pollution research planning seminar*, Washington (Public Health Service), p. 32

Logan, W. P. D. (1949) *Lancet*, **1**, 78

Logan, W. P. D. (1953) *Lancet*, **1**, 336

Logan, W. P. D. (1956) *Lancet*, **1**, 722

Looney, W. B., Hasterlik, R. J., Brues, A. M. & Skirmont, E. (1955) *Amer. J. Roentgenol.*, **73**, 1006

Lorenz, E. & Congdon, C. C. (1954) *Ann. Rev. Med.*, **5**, 323

Los Angeles Air Pollution Control District (1951) *Second technical and administrative report on air pollution control in Los Angeles County, 1950-51*, Los Angeles

Los Angeles Air Pollution Control District (1954) *Progress in controlling gasoline vapors*, Los Angeles (Publication No. 46-A)

Lynch, F. W. (1951) *A.M.A. Arch. Derm. Syph.*, **63**, 503

McCabe, L. C. (1951) *Industr. Engng Chem.*, **43**, 79a

McCabe, L. C., ed. (1952) *Air pollution. Proceedings of the United States Technical Conference on...*, New York

McCabe, L. C. (1954) *Industr. Engng Chem.*, **46**, 1646

McCabe, L. C. & Clayton, G. D. (1952) *A.M.A. Arch. industr. Hyg.*, **6**, 199

McConnell, R. B., Gordon, K. C. T. & Jones, T. (1952) *Lancet*, **2**, 651

McCord, C. P. & Witheridge, W. N. (1949) *Odors, physiology and control*, New York

McCully, C. R., Fisher, M., Langer, G., Rosinski, J., Glaess, H. & Werle, D. (1956) *Industr. Engng Chem.*, **48**, 1512

McEwen, W. K. (1956) In: *Proceedings of the air pollution research planning seminar*, Washington (Public Health Service), p. 11

Macgregor, A. (1947) *Med. Offr.* **78**, 157
Machle, W., Beyer, E. C. & Gregorius, F. (1948) *Occup. Med.*, **5**, 671
Machle, W. & Gregorius, F. (1948) *Publ. Hlth Rep. (Wash.)*, **63**, 1114
Mage, J. & Batta, C. (1932a) *Chim. et Ind.*, **27**, 961
Mage, J. & Batta, G. (1932b) *Chim. et Ind.*, **30**, 787
Magill, P. L. (1949) *Industr. Engng Chem.*, **41**, 2476
Mancuso, T. F. (1951) *Industr. Med. Surg.*, **20**, 393
Mancuso, T. F. & Hueper, W. C. (1951) *Industr. Med. Surg.*, **20**, 358
Marinelli, L. D., Quimby, E. H. & Hine, C. J. (1948) *Amer. J. Roentgenol.*, **59**, 260
Marshall, W. R. L. (1952) *A century of London weather*, London
Mayer, R. L. (1929) *Arch. Derm. Syph. (Berl.)*, **158**, 266, 734
Mayer, R. L. & Foerster, M. (1929) *Zbl. Gew.-Hyg.*, **6**, 171
Meneely, G. R. (1957) *Dis. Chest*, **31**, 125
Miller, C. A., Davis, M. W., Goldman, A. & Wyatt, J. P. (1953) *Arch. industr. Hlth*, **8**, 453
Miller, W. R., Jarrett, E. T., Willman, T. L., Hollaender, A., Brown, E. W., Lewandowski, T. & Stone, R. S. (1948) *J. infect. Dis.*, **82**, 86
Mills, C. A. (1943) *Amer. J. Hyg.*, **37**, 131
Mills, C. A. (1945) *R. I. med. J.*, **28**, 496
Mills, C. A. (1946a) *Canad. Hosp.*, **23**, 47
Mills, C. A. (1946b) *Cincinn. med. J.*, **27**, 441
Mills, C. A. (1946c) *Cincinn. med. J.*, **27**, 624
Mills, C. A. (1949a) *Hygeia*, **27**, 684
Mills, C. A. (1949b) *J. Amer. med. Ass.*, **140**, 556
Mills, C. A. (1950) *Science*, **111**, 67
Mills, C. A. & Mills-Porter, M. (1948) *Occup. Med.*, **5**, 614
Mills, K. D. & Seipp, G. A. (1955) *Air Pollution Foundation Report*, No. 8. Los Angeles, p. 1
Mitchell, C. W. & Davenport, S. J. (1924) *Publ. Hlth Rep. (Wash.)*, **39**, 1
Molesworth, E. H. (1927) *Med. J. Aust.*, **1**, 878
Morgan, G. W. (1949) *Nucleonics*, **4**, 24
Moritz, A. R. & Henriques, F. W. (1952) *J. Lab. Invest.*, **1**, 167
Morris, G. E. (1956) *A.M.A. Arch. industr. Hlth*, **13**, 480
Moshman, J. (1951) *J. appl. Physiol.*, **4**, 145
Mountin, J. W., Dorn, H. F. & Boone, B. R. (1939) *Publ. Hlth Rep. (Wash.)*, **54**, 1255
Mueller, F. H. (1939) *Z. Krebsforsch.*, **49**, 57
Muller, H. J. (1955) *Bull. atom. Scient.*, **11**, 210
National Academy of Sciences (1956) *The fluorosis problem in livestock production*, Washington (Publication No. 381)
Nature (Lond.), 1909, **81**, 468
Nielsen A. & Clemmesen, J. (1954) *Dan. med. Bull.*, **1**, 194
Niskanen, K. O. (1949) *Acta path. microbiol. scand. (Suppl.)*, **80**, 1
Noro, L. (1945) *Nord. Med.*, **20**, 771
Oberling, C. (1954) *Presse méd.*, **62**, 712
Ochsner, A., DeCamp, P. T., DeBakey, M. E. & Ray, C. J. (1952) *J. Amer. med. Ass.*, **148**, 691
Opie, E. L. (1921) *Proc. Smoke Prev. Ass.*, p. 16
Pancheri, G. (1955) In: Mallette, F. S., ed. *Problems and control of air pollution*, New York, p. 253
Parker, A. (1950) *J. roy. Soc. Arts*, **99**, 85
Parker, A. (1957) *Amer. J. publ. Hlth*, **47**, 559
Pattle, R. E. & Cullumbine, H. (1956) *Brit. med. J.*, **2**, 913
Peller, S. (1939) *Hum. Biol.*, **11**, 130
Peller, S. & Pick, P. (1952) *Amer. J. med. Sci.*, **224**, 154
Pemberton, J. & Goldberg, C. (1954) *Brit. med. J.*, **2**, 567

Phillips, A. J. (1954) *Canad. med. Ass. J.*, **71**, 242

Pinto, S. S., Petronella, S. J., John, D. R. & Arnold, M. F. (1950) *Arch. industr. Hyg.*, **1**, 437

Policard, A. (1949) *Rev. Méd. min.*, No. 8, p. 7

Policard, A. & Collet, A. (1952) *Arch. industr. Hyg.*, **5**, 527

Portheine, F. (1954) *Arch. Gewerbepath. und Gewerbehyg.*, **13**, 253

Pott, P. (1775) *Chirurgical observations relative to the cataract, the polypus of the nose, the cancer of the scrotum, the different kinds of ruptures, and the mortification of the toes and feet*, London, p. 63

Publ. Hlth Bull. (Wash.), 1926, No. 163

Publ. Hlth Rep. (Wash.), 1930, **45**, 2032

Ravina, A. (1954) *Presse méd.*, **62**, 881

Regan, C. J. (1932) *Chem. & Ind.*, **51**, 605

Reid, L. (1956) *Proc. roy. Soc. Med.*, **49**, 771

Renzetti, N. A. & Gobran, V. (1957) *Studies of eye irritation due to Los Angeles smog, 1954-1956*, Los Angeles (Air Pollution Foundation)

Riazonov, V. A. (1954) [*Sanitary safeguarding of atmospheric air*], Moscow

Rich, H. M. (1924) *Trans. Amer. clin. climatol. Ass.*, **40**, 331

Roffo, A. H. (1933) *Amer. J. Cancer*, **17**, 42

Roholm, K. (1936) *Hospitalstidende*, **79**, 1337

Roholm, K. (1937) *Fluoride intoxication*, London

Roholm, K. (1938) *J. industr. Hyg.*, **19**, 126

Romanoff, A. (1939) *J. Allergy*, **10**, 166

Ronge, H. E. (1948) *Acta physiol. scand. (Suppl. 49)*, **15**, 1

Roueché, B. (1950) *The New Yorker*, **26**, (31), 33

Rowher, S. A. & Haller, H. L. (1950) *J. Amer. med. Ass.*, **144**, 104

Russell, W. T. (1924) *Lancet*, **2**, 335

Russell, W. T. (1926) *Lancet*, **2**, 1128

Sadowsky, D. A., Gilliam, A. G. & Cornfield, J. (1953) *J. nat. Cancer Inst.*, **13**, 1237

Safety Engng, 1946, **92**, 56

Salk, J. E. (1944) *J. Immunol.*, **49**, 87

Sayers, R. R. & Davenport, S. J. (1937) *Publ. Hlth Bull. (Wash.)*, No. 195

Sayers, R. R., Fieldner, A. C., Yant, W. P., Thomas, B. G. H. & McConnell, W. J. (1924) *Exhaust gases from engines using tetraethyl of lead gasoline*, Washington (United States Bureau of Mines Report of Investigations 2661)

Schnurer, L. (1938) *J. industr. Hyg.*, **20**, 14

Schrader, J. H., Coblentz, M. H. & Korff, F. A. (1929) *Amer. J. publ. Hlth*, **19**, 717

Schrek, R., Baker, L. A., Ballard, G. P. & Dolgoff, S. (1950) *Cancer Res.*, **10**, 49

Schrenk, H. H., Heimann, H., Clayton, G. D., Gafafer, W. M. & Wexler, H. (1949) *Air pollution in Donora, Pennsylvania*, Washington (Public Health Service Bulletin No. 306)

Scott, J. A. (1953) *Publ. Hlth Rep. (Wash.)*, **68**, 474

Selye, H. (1957) *The stress of life*, New York

Senn, C. L. (1948) *Amer. J. publ. Hlth*, **38**, 962

Shapiro, M. P., Keen, P., Cohen, L. & Murray, J. F. (1953) *Brit. J. Cancer*, **7**, 45

Shaw, N. & Owens, J. S. (1925) *The smoke problems of great cities*, London

Shilen, J. (1949) *Industr. Med.*, **18**, 70

Shilen, J., Mellor, J. F., Stang, A. M., Sharrah, J. S. & Robson, C. D. (1949) *The Donora smog disaster*, Harrisburg, Penn. (Pennsylvania Department of Health)

Sievers, R. F., Edwards, T. I., Murray, A. L. & Schrenk, H. H. (1942) *J. Amer. med. Ass.*, **118**, 585

Silson, J. E., Kleinfeld, M. & Harris, S. J. (1955) *A.M.A. Arch. industr. Hlth*, **12**, 503

Silverberg, M. G. & Heimann, H. (1941) *J. invest. Derm.*, **4**, 193

Silverman, A. (1929) *Heating, Piping & Air Conditioning*, **1**, 604

Skipper, H. E., Nolan, C. & Simpson, L. (1951) *J. biol. Chem.*, **189**, 159

Smithard, E. H. R. (1954) *Monthly Bull. Minist. Hlth Lab. Ser.*, **13**, 26

Snegireff, L. S. & Lombard, O. M. (1951) *A.M.A. Arch. industr. Hyg.*, **4**, 199

Snell, F. M. & Neel, J. V. (1949) *Arch. int. Med.*, **84**, 569

Spitz, S., Maguigan, W. H. & Dobriner, K. (1950) *Cancer (Philad.)*, **3**, 789

Stanford Research Institute (1948) *The smog problem in Los Angeles County ; first interim report*, Los Angeles

Stanford Research Institute (1949) *The smog problem in Los Angeles County ; second interim report*, Los Angeles

Stanford Research Institute (1950) *The smog problem in Los Angeles County ; third interim report*, Los Angeles

Stanford Research Institute (1954) *The smog problem in Los Angeles County*, Los Angeles

Steel, M. & Feltham, D. V. G. (1950) *Lancet*, **1**, 108

Stern, A. C. (1945) *Heating & Ventilating*, **42**, 73

Sterner, J. H. & Eisenbud, M. (1951) *A.M.A. Arch. industr. Hyg.*, **4**, 123

Sterner, P. E. (1954) *Cancer Res.*, **14**, 103

Stocks, P. & Campbell, J. M. (1955) *Brit. med. J.*, **2**, 923

Stokinger, H. E. (1956) In: *Proceedings of the air pollution research planning seminar*, Washington (Public Health Service), p. 21

Stoner, J. B. (1913) *Milit. Surg.*, **32**, 372

Sumari, P. (1949) In: *Proceedings of the ninth international congress on industrial medicine*, London, p. 603

Sumerford, W. T., Hayes, W. J., Johnston, J. M., Walter, K. & Spillane, J. (1953) *A.M.A. Arch. industr. Hyg.*, **7**, 383

Symanski (1942) *Dtsch. med. Wschr.*, **68**, 192

Taplin, G. V., Buettner, H., Gautschi, M. L. & Johnson, M. J. (1951) *Stanford med. Bull.*, **9**, 112

Tebbens, B. D., Thomas, J. F. & Mukai, M. (1956a) *A.M.A. Arch. industr. Hlth*, **13**, 567

Tebbens, B. D., Thomas, J. F. & Mukai, M. (1956b) *A.M.A. Arch. industr. Hlth*, **14**, 413

Tipton, I. H. (1956) *Spectrographic procedure of analysis of human tissues* (mimeographed), Knoxville, Tenn. (University of Tennessee)

Tipton, I. H., Steiner, R. L., Foland, W. D., Mueller, J. & Stanley, M. (1957) *Spectrographic analysis of the tissues from autopsies of 24 instantaneous deaths* (mimeographed) Knoxville, Tenn. (University of Tennessee)

Tonney, F. O., Hoeft, G. L. & Somers, P. (1930) *J. prev. Med.*, **4**, 139

Tsuzuki, M. (1955) *Münch. med. Wschr.*, **97**, 988

Turk, A. (1954) *American Society of Testing Materials, Publication* No. 164, New York

United States, Department of Health, Education, and Welfare, Public Health Service (1953) *Health of workers in chromate producing industry, study*, Washington

United States, Department of Health, Education, and Welfare, Public Health Service (1956) *Clinical memoranda on economic poisons*, Washington

United States, National Bureau of Standards (1953) *Maximum permissible amounts of radioisotopes in the human body and maximum permissible concentrations in air and water*, Washington (National Bureau of Standards Handbook 52)

Valentine, H. (1936) *Plomb tétra-éthyle et hygiène industrielle*, Paris

Van Wijk, A. M. & Patterson, H. W. (1940) *J. industr. Hyg.*, **22**, 31

Voegtlin, C. & Hodge, H. (1949) *Pharmacology and toxicology of uranium compounds*, New York

Waller, R. E. (1950) In: *Proceedings of the National Smoke Abatement Society, 17th Annual Conference*, London, p. 53

Waller, R. E. (1952) *Brit. J. Cancer*, **6**, 8

Wallin, J. E. W. (1913) *Psychological aspects of the problem of atmospheric smoke pollution*, Pittsburgh (Mellon Institute Smoke Investigation Bulletin No. 3)

Walpole, H. (1955) Cited by Cullumbine, H., Pattle, R. E. & Burgess, F. (1955) *Toxicity of fog*. In: *7e Congrès international de Pathologie comparée*, Lausanne, p. 111

Warren, S. (1942) *Arch. Path. (Chicago)*, **34**, 443, 562, 749, 917, 1070

Warren, S., ed. (1956) *Pathologic effects of atomic radiation*, Washington (National Academy of Sciences)

Watkinson, E. A. (1955) In: Mallette, T. S., ed. *Problems and control of air pollution*, New York, p. 69

Weaver, E. R. (1950) *Control of odors*, Washington (United States National Bureau of Standards Circular No. 491)

Weedon, F. R., Hartzell, A. & Setterstrom, C. (1939) *Contr. Boyce Thompson Inst.*, **10**, 281

Wells, W. F. (1955) *Airborne contagion and air hygiene*, Cambridge, Mass.

Whisler, B. A. (1940) *J. Science (Iowa St. Coll.)*, **14**, 215

Wilkins, E. T. (1954) *J. roy. sanit. Inst.*, **74**, 1

Wilson, E. D., Gates, I., Owen, H. R. & Dawson, W. T. (1926) *J. Amer. med. Ass.*, **87**, 319

Wilson, I. B. & La Mer, V. K. (1948) *J. industr. Hyg.*, **30**, 265

Winslow, C.-E. A. & Herringon, L. P. (1935) *Heating, Piping & Air Conditioning*, **7**, 551

Wright, B. M. (1950) *Nature (Lond.)*, **166**, 538

Wynder, E. L. (1952) *New Engl. J. Med.*, **246**, 492, 538, 573

Wynder, E. L. & Graham, E. A. (1950) *J. Amer. med. Ass.*, **143**, 329

Yamasaki, F. & Kakehi, K. (1954) *Kagaku*, vol. 24

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EFFECTS OF AIR POLLUTION ON ANIMALS

Introduction

Interest in the effects of air pollution on animals has generally developed as a corollary to the concern about this influence on human health. Studies in which animals have been exposed experimentally to air pollutants have been prompted by the need for specific information concerning the toxicity of various known pollutants. Such information will assist in defining the effects on man. Moreover, animal morbidity and mortality following air pollution disasters have been recorded, at least partially, to seek a better understanding of the human injury.

There has been a direct concern about the injurious effects of air pollution on animals because of the considerable economic loss which has been claimed in some instances and is potentially associated with this hazard. This interest is illustrated by the research which has been devoted to the problem created by airborne fluorides.

The concern over the threat of air pollution to the health of man and animals has developed for the most part during the past decade. With the exception of the reports dealing with fluorosis, there is little to be found in the veterinary literature about air pollution as a cause of disease. Most of the information concerning the natural exposure of animals to air pollution is contained in the reports of some major air pollution disasters—e.g., Donora, London, Poza Rica. Recently, considerable information has been reported from medical research laboratories which describes the results of experimental exposure of small animals to various air pollutants. The advent of nuclear fission has added a new category of pollution to be studied and evaluated.

This chapter will attempt to appraise the reports which developed from investigations of the air pollution disasters at Donora, London and Poza

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Rica. Attention will also be given to the laboratory reports concerning air pollutants to which animals have been naturally exposed. The information concerning animal fluorosis will be reviewed, as will observations pertaining to the effects of radioactive fall-out on animals.

Major Air Pollution Disasters

A detailed investigation of both human and animal injury was conducted following the air pollution incident at Donora, Pennsylvania in 1948 (*Publ. Hlth Bull. (Wash.)*, 1949). A retrospective survey disclosed that an appreciable number of animals were reported to have become ill and that some died during the week of intense smog. The information concerning animal morbidity and mortality was obtained from the owners of both pets and farm animals by personal interviews conducted by registered nurses among the urban population and by a veterinarian among the farmers. Reports obtained by the nurses were subsequently investigated by the veterinarian.

Corroborative evidence was sought through conferences with three local veterinarians, technicians in a local dairy-cattle breeding association, two county agents, three local poultry dealers and a slaughterhouse operator. Four retail milk plants in the area were visited to examine milk production records during and after the smog. This group of observers was unable to indict the intense smog as being responsible for any noticeable effect upon the health or performance of animals. Milk production did not decline during nor immediately after the smog, according to the dairy operators.

The reports obtained through interviews with animal owners and caretakers indicated that the different species varied in their susceptibility to injury. Dogs were reportedly the most susceptible species. Of 229 dogs included in the survey, 15.5% were reported as having become ill and 10 canine deaths were attributed to the smog. Nearly all of these fatalities occurred among dogs of less than one year of age.

The investigators grouped the signs of the canine illnesses attributed to air pollution into three syndromes. Signs of the respiratory syndrome, which occurred most frequently, included cough, sneezing, conjunctival congestion, dyspnoea and nasal discharge. Signs of the digestive syndrome included emesis, retching and diarrhoea. The third syndrome consisted of anorexia which occurred with or without lassitude. With the exception of the fatal cases, the canine illnesses were generally mild and short-lasting. The mean duration of the illnesses reported for 31 dogs was between three and four days.

Twelve of 165 cats which were included in the animal investigation at Donora were reported as having become sick during the smog interval.

Three cats died after developing signs which were similar to those described in dogs.

There were few observations of illness and death among poultry. Mild signs of respiratory distress were reported as occurring in two of 43 urban flocks. Four rural poultrymen observed that their chickens became sick and that approximately 40 % of the affected birds had died.

The larger farm animals including horses, cattle, sheep and swine were generally unaffected by the Donora smog. Three dairy farmers reported, however, that their cows had developed a cough which coincided with the occurrence of intense air pollution. At one farm five cases of calf pneumonia were said to have occurred immediately after the smog.

In appraising the reports of animal injury during the intense smog at Donora several considerations appear to be important. It is significant that professional and experienced observers of animal health failed to regard the smog as a factor in the morbidity and mortality which occurred among animals at that time. It should also be noted that while it is entirely possible that the reported illnesses could have been caused by the inhalation of irritating gases and fumes, the signs which were observed are consistently associated with common animal diseases occurring in this locality.

The significance of the animal effects reported at Donora is further clouded by the inability to indict specific air pollutants as likely causes for the morbidity and mortality which occurred. The chief contaminants of the Donora smog were later estimated to have been well within the range of their respective toxic levels. The farm animals and poultry were not located in the centre of intense pollution and so relative exposures are unknown.

It is impossible to make a fair estimate of the relative susceptibility of man and animals in this episode. Although the reactions of the animals are not biased by subjective reflection, the reports of the owners may be. In man, 10 % were severely affected and an additional 17 % moderately affected. The 15 % of dogs reported as having become ill might have consisted only of those with rather severe reactions, making the dogs' susceptibility roughly equivalent to that of man. It seems unlikely that any of the species of animals affected at Donora would provide a sensitive biological indicator for the degree or nature of human illness caused by this type of air pollution.

During the interval of the London fog in 1952, a number of prize cattle were reported as having been severely affected (*Brit. med. J.*, 1953; Joules, 1954). These animals, which were grouped for exhibition, developed acute respiratory distress. Five of them died, 11 were subjected to emergency slaughter and over 40 others developed signs which were attributed to the fog. Post-mortem examinations revealed acute bronchiolitis, emphysema and right heart failure. Sulfur dioxide was the chemical component of the

London fog which was proposed as the cause for the morbidity and mortality (Drinker, 1953).

A related sulfur compound, hydrogen sulfide, has been held responsible for another major air pollution disaster at Poza Rica, Mexico, 1950 (McCabe & Clayton, 1952). Following this incident, an investigation disclosed that the animal population apparently had been affected. An undetermined number of canaries, chickens, cattle, pigs, geese, ducks and dogs were exposed to the polluted atmosphere at Poza Rica. The reports of animal injury during this disaster disclosed that 100 % of the canaries in the area had died. It was estimated that 50 % of the other exposed animals had died during the period of air pollution.

Exposure of Laboratory Animals

A considerable body of information is beginning to become available from laboratory experimentation with small animals. Such experimentation is capable of providing more specific and more reliable information in three basic categories. First, the animals themselves can be of known genetic constitution, with relatively well defined physiological constants and susceptibilities to infectious disease (Russell, 1955) and, eventually, to air pollutants. Secondly, the exposures to air pollutants may be controlled both qualitatively and quantitatively. Thirdly, responses of the animals to the exposures may be measured in detail and by techniques not readily applicable to man. Laboratory experiments serve both in the basic field of testing known contaminants and as a follow-up to field epidemiological studies, to determine whether true causal relationships exist between potential causes and effects noted in the field.

Most available results from laboratory experiments define the reactions of animals to substantial concentrations of specific chemical air pollutants. The study of potential health hazards from air pollution as it occurs today must be concerned more with chronic low-level exposure than with the acute episode of short duration and high concentration. Although further acute episodes may be expected and anticipated, and may even produce local disasters of consequence, it is the long-term, widespread exposure to daily air pollution which must be examined thoroughly for delayed effects upon the health of animals and man. Epidemiological investigation of " natural " pollution may be so confounded by environmental factors and other intercurrent pathology that no specific effects can be differentiated. It is upon this thesis that the need for experimental animal epidemiology is predicated.

Animal species have been shown to vary in their susceptibility to various air pollutants. The observations at Poza Rica suggest that avian species, especially canaries, may be considerably more susceptible to hydrogen sulfide than are the more common laboratory animals. The fate of fattened cattle during the London fog elicits the conjecture that increased stress associated with the rapid deposit of considerable fat may alter the cardiorespiratory functions, thereby increasing susceptibility to air pollutants.

Rats exposed to silica and feldspar dust did not exhibit increased susceptibility to lobar pneumonia (Baetjer & Wintinner, 1944). Comprehensive studies have been made of the effects of sulfur dioxide and sulfuric acid on guinea-pigs (Amdur, 1954; Comar et al., 1957). A combination of these two had a much more marked effect than equivalent concentrations of either alone. Effects were demonstrated on growth, lung pathology, and airway resistance. Similar physiological effects have been noted in man (Amdur, Melvin & Drinker, 1953; Greenwald, 1954), and similar signs of illness and post-mortem pathology were noted in the cattle exposed to the London fog (Pattle & Cullumbine, 1956). Hyperpnoea, dyspnoea, and depression are exhibited commonly by most laboratory animals. Guinea-pigs are recorded as being more susceptible to these toxins than mice, rats or goats (Pattle & Cullumbine, 1956). Necropsies of exposed guinea-pigs have revealed the presence of pulmonary haemorrhages, oedema, and, in addition, consolidation and hepatization (Amdur, Schulz & Drinker, 1952). Studies in progress in this same laboratory show that the combination of sulfur dioxide and a sodium chloride aerosol produce airway resistance effects many times greater than sulfur dioxide alone.

At this time a number of studies are well under way to determine the effects of single or mixed air pollutants on the enzyme systems of specified tissues or cells. Preliminary reports of a number of these projects are included in the proceedings of the Air Pollution Research Planning Seminar (US Department of Health, Education, and Welfare, 1956).

Exposure to Oxidizing Air Pollution

The natural exposure of animals to atmospheric pollution characterized by the reaction of unsaturated hydrocarbons and oxides of nitrogen in the presence of sunlight has been investigated but superficially to date. This type of air pollution is exemplified by that which mars the atmosphere of Los Angeles (Haagen-Smit & Fox, 1956).

In 1954, an intensive survey of the air pollution problem in Los Angeles was conducted. During this time the effects on animal health were cursorily investigated. The information obtained was apparently regarded as inconclusive since it was not included in the published report of the survey (California State Department of Public Health, 1955). More recently, it has been discovered through the medium of a questionnaire that veterinarians in Southern California occasionally incriminate air pollution as a cause of illness among animals (Catcott, 1957, unpublished data). Signs of ocular and upper respiratory tract irritation among dogs and pet birds are rather frequently attributed to atmospheric contaminants.

Artificial exposure of laboratory animals has provided some evidence of the effects caused by some of the chemical agents present in this type of smog. The studies of the carcinogenic properties of ozonized hydrocarbons illustrate this information (Kotin, 1956). Both dermal and pulmonary cancers have been produced in mice artificially exposed to an irradiated mixture of ozone and unsaturated hydrocarbons. Skin painting with aromatic hydrocarbons produced skin tumours in both C57 black and strain-A mice (Kotin, Falk & Thomas, 1956). Skin painting with aliphatic hydrocarbons also produced skin tumours in C57 black mice. Of more interest and probably more significance is the finding that pulmonary tumours were produced in strain-A mice after their exposure to an atmosphere of ozonized gasoline (Kotin & Falk, 1956) In these mice tumours developed in 41 % under washed air conditions and in 80 % in polluted air. Results on the C57 black mice under similar exposure are reported by Kotin & Falk (1958). The control animals showed a very low percentage of lung tumours, whereas over one-third of those exposed to polluted air produced tumours. Additional biological effects on these mice will be reported in detail. At the moment it has been noted that the mice housed in a polluted atmosphere showed a consistent weight deficit in comparison with the controls.

These laboratory observations are at present being complemented with a field survey of pulmonary pathology in dogs (Catcott, McCammon & Kotin, 1958). In this study, dogs of known age and period of residence in the Los Angeles area are being examined post mortem. Both gross and microscopic observations are being made of the major organs, particular attention being given to pulmonary tissues.

Ozone was considered to be an important component of the Los Angeles type of air pollution. Rather extensive studies on the toxicology of ozone have been conducted and are still being pursued (Stokinger, 1954; Stokinger, Wagner & Wright, 1956; US Department of Health, Education and Welfare, 1956). The irritant qualities of ozone have been demonstrated in dogs, cats, rabbits, mice and guinea-pigs. Exposures at toxic levels have produced marked pulmonary changes, including death. Oedema and haemorrhage characterize the morphological changes of lungs which have been injured with ozone. In nearly all experiments, the concentrations of ozone have been greater than those observed to occur naturally. Although ozone was considered to be a representative constituent of the oxidizing type of smog, opinion is now developing that ozone itself is not the responsible agent in this type of smog. Experiments in progress indicate that animals may develop a tolerance to ozone, or that they may be made more susceptible through the medium of environmental stress.

Animal Fluorosis

While fluorine toxicosis is usually induced by a less direct process than inhalation, the chemical agent is air-borne. Fluorosis of animals has become fairly well defined during the past ten years. Chronic fluoride poisoning probably results from the distribution of fluorine in the air. Fluorides are widely distributed in soils, water and animal feeds. It has been stated that it would be difficult to devise an animal ration which would contain less than 2-3 p.p.m. of fluorides (Phillips, 1952).

The sources of fluoride present in, or on, pasture crops include: (*a*) the natural dusts from soil in certain localities. Some top soil in Idaho has been found to contain as much as 1640 p.p.m. of fluoride; (*b*) dusts and gases from certain factories; (*c*) the combustion of coal, which results in the dispersion of fluoride-containing materials (Largent, 1952). Gaseous effluents from the enamelling, cement, and aluminium industries and from cryolite or acid phosphate production contain hydrogen fluoride or other gaseous fluorides.

The occurrence and distribution of livestock fluorosis in the USA has been contemporaneous with the development of the industries previously mentioned. The fluoride-containing effluents from these industries may contaminate the forage crops which are subsequently ingested by animals. Air contamination is greatest in the area adjacent to industrial sources and decreases with distance as the effluents become dispersed (Huffman, 1952). Sufficient forage contamination to produce the early signs of fluorine toxicosis may occur at a distance of seven miles (11 km) or more in the direction of the prevailing wind. Certain industrial establishments have made every effort to prevent this type of contamination; others are still a problem.

Fluorine is a protoplasmic poison. It has a marked affinity for calcium and interferes with normal calcification (Madsen, 1942). Livestock are observed to be more resistant than are humans to dental mottling, which is an early sign of fluorine toxicosis. Cattle and sheep are the most frequently affected animals (Largent, 1952). Experimental work has indicated that the maximum safe levels of daily fluoride ingestion are: 3 mg/kg bodyweight for cattle and sheep, and 10 mg/kg for chickens. Only one clinical case of fluorine poisoning in a horse has been reported in the USA (Largent, 1952).

The pathogenesis of fluorine toxicity consists of hypoplasia of dental enamel followed at higher levels of fluorine intake by abnormal growth of bones. The pathology is manifested by mottling, staining and pitting of incisor teeth. Generally, only the permanent teeth are affected. Excessive wearing of incisor and molar teeth with subsequent gingival injury may occur at high levels of fluorine intake. Bone lesions may develop at any age. Their appearance indicates that there has been a high fluorine intake over a long period. The pathology is manifested by isolated exostoses or by a diffuse thickening of long bones. Lameness is a consequence when joint surfaces become involved. Signs of advanced fluorosis include anorexia, diarrhoea, weight loss, lowered fertility and reduced milk production. Positive diagnosis of fluorosis is based on a chemical determination of fluoride levels in bones or teeth. Normal fluoride levels in bone ash of unaffected animals range from 200 to 800 p.p.m. About 2 mg daily of fluorine per kg of bodyweight will produce dental mottling in cattle within four to five years. The toxicity of fluorine appears to be greater in forage contaminated with gaseous effluents than with rock phosphate. The storage of fluorine in the soft tissues of cows on high level intakes is reportedly low. Therefore, neither meat nor milk from such animals is likely to be injurious to humans. The prospect of controlling industrial fluorosis of livestock appears to be good. The element is so widely distributed in nature, however, that its complete eradication as an industrial waste is unlikely (Huffman, 1952).

The exposure of rabbits and guinea-pigs to high concentrations of hydrogen fluoride has been observed to be irritating to the respiratory tract (Machle et al., 1934). Slowing of the respiratory rate was observed at low concentrations. Increased exposure caused corneal erosion, necrosis of turbinate bones and marked conjunctival and nasal discharges. Death of these laboratory animals following their exposure to hydrogen fluoride was usually due to bronchopneumonia.

Exposure to Ionizing Radiation

Radioactive fall-out from nuclear weapons testing is a unique air pollutant in that the associated biological effects occur as a result of ionizing radiation rather than chemical toxic action. The effects of radiation on animals are similar qualitatively to those in humans (Hollaender, 1954; National Academy of Sciences, 1956a). They can arbitrarily be categorized as acute radiation effects and the delayed or long-term effects.

Signs of acute radiation injury develop within a period of hours to weeks following exposure. Only fall-out occurring close to the weapon's test site can produce radiation levels sufficiently high to result in acute radiation effects (Cronkite, Bond & Dunham, 1955; National Academy of Sciences, 1956b; US Department of the Army, 1957).

Our primary interest is in the long-term effects that may be associated with global fall-out. The most important long-term effects to consider are: (*a*) cancer (including leukaemia); (*b*) shortening of life span (non-specific aging); and (*c*) the genetic or mutation effect. These effects generally require several years, or, in the case of genetic changes, generations following exposure, to be manifested. Our concern about these effects in animals is not only in the interests of animal health, but principally for the information that can be extrapolated to man. In the USA, the National Academy of Sciences (1956a) points out:

> Laboratory studies of long-term effects have been relatively few, partly because their importance was not early appreciated and partly because they are very expensive and time consuming owing to the necessity for maintaining considerable numbers of animals for all, or most, of their normal life spans. In consequence, the data on late effects in animals are meagre in certain areas. Such data as there are in man are sufficiently in agreement with those on other mammals to lead to the expectation that extrapolation from lower mammals to man will be possible with fair accuracy.

The Academy (1956b) also reports:

> Radiation from fall-out inevitably contaminates man's food supply. Radioactive elements in the soil are taken up and concentrated by plants. The plants may be eaten by humans, or by animals which in turn serve as human food. At present the contamination is negligible. But the maximum tolerable level is not known. There is not nearly enough information about the long-term biological effects on man or animals from eating radiation-contaminated food. Research in this area is urgently needed.

We are interested in the quantities of radioactive fission products being assimilated by animals throughout the world, because such data provide a biological index of the level of environmental contamination and give warning of the build-up of potentially hazardous levels.

Compared to other air pollutants it is relatively easy to determine environmental levels of radioactive fall-out and to measure the body burden of fission products assimilated by animals. In recent years there have been numerous studies to measure levels of select fission products naturally occurring in animal tissues and animal products as a result of fall-out (Anderson et al., 1957; Comar et al., 1957; Kulp, Eckelmann & Schulert, 1957; Van Middlesworth, 1956).

Summary

Reports concerning the effects of air pollution on the health of animals have four general sources. Reports of animal injury and death have followed investigations of the air pollution disasters at Donora, London, and Poza Rica. Extensive studies of air-borne fluorides and their relation to animal health are available. Observations of the effects of air-borne radioactive substances on animals provide a third source. Finally, the reports of artificial exposure of laboratory animals have enlarged our understanding of the effects of various air pollutants on animals.

It must be conceded that the present information concerning this subject is quite inadequate. The reports of animal morbidity and mortality which followed major air pollution episodes should be regarded critically. The investigations of these acute and intense exposures to air pollution have been done retrospectively. It is significant that the owners' reports of injury to animals could not be corroborated by professional observers at the Donora disaster. The high rate of animal mortality which allegedly occurred at Poza Rica is generally in contradiction to the information concerning the relative susceptibility to air pollutants of animal species which have been studied experimentally.

The synergistic roles of physiological and of external environmental influences on reactions to air pollution indicate that the interactions of many factors may be necessary to produce critical situations. Genetic attributes of the individual animal as well as the species may define specific parameters of physiology and nutrition within the animal, whereas meteorology and type source of air pollution define exposures. The association of animals and atmosphere may be the final requirement for specific biological effects.

There has been very little study of animals subjected to natural exposure to air pollution of the kind prevalent on the west coast of the USA. Investigations of naturally occurring effects of this form of air pollution are at present being conducted. Most of our information concerning the oxidant type of pollutant has been and currently is being obtained in experimental studies of laboratory animals.

In contrast to the paucity of information concerning natural exposure to most air-borne pollutants, the effects of fluorides on animals have been defined well. The incidence of animal fluorosis and its nature have been described. Methods of controlling this hazard to animal health are known and utilized when the threat of fluorosis occurs.

Laboratory research has provided important information concerning the effects of specific pollutants on animals. Mice, rabbits, guinea-pigs, rats and monkeys have been utilized to demonstrate the toxic properties of such air pollutants as sulfur dioxide, sulfuric acid, hydrogen sulfide, ozone, nitrogen dioxide, organic compounds, and some dusts. Information which has been obtained by artificial exposure of animals is providing some indices of both human and animal effects to be expected from natural exposures. A well-integrated attack, in the field and in the laboratory, will be necessary to divulge the true details of the biological effects of polluted air.

REFERENCES

Amdur, M. O. (1954) *Publ. Hlth Rep. (Wash.)*, **69**, 503
Amdur, M. O., Melvin, W. W. & Drinker, P. (1953) *Lancet*, **2**, 758
Amdur, M. O., Schulz, R. Z. & Drinker, P. (1952) *A.M.A. Arch. industr. Hyg.*, **5**, 318
Anderson, E. C., Schuch, R. L., Fisher, W. R. & Langham, W. (1957) *Science*, **125**, 1273
Baetjer, A. M. & Wintinner, F. J. (1944) *J. industr. Hyg.*, **26**, 101

Brit. med. J., 1953, **1**, 321

California State Department of Public Health (1955) *Clean air for California*, San Francisco

Catcott, E. J., McCammon, C. J. & Kotin, P. (1958) *J. Amer. vet. med. Ass.*, **133**, 331

Comar, C. L., Trum, B. F., Kuhn, U. S. G., III, Wasserman, R. H., Nold, M. M. & Schooley, J. C. (1957) *Science*, **126**, 16

Cronkite, E. P., Bond, V. P. & Dunham, C. L. (1956) *Some effects of ionizing radiations on human beings*, Washington (US Atomic Energy Commission, TIC 5358)

Drinker, P. (1953) *A.M.A. Arch. industr. Hyg.*, **7**, 275

Greenwald, I. (1954) *A.M.A. Arch. industr. Hyg.*, **10**, 455

Haagen-Smit, A. J. & Fox, M. M. (1956) *Industr. Engng Chem.*, **48**, 1484

Hollaender, A., ed. (1954) *Radiation biology*, New York, Vol. 1, pt 2

Huffman, W. T. (1952) *Effects on livestock of air contamination caused by fluoride fumes.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 59

Joules, H. (1954) *Brit. med. J.*, **2**, 1259

Kotin, P. (1956) *Cancer Res.*, **16**, 375

Kotin, P. & Falk, H. L. (1956) *Cancer (Philad.)*, **9**, 910

Kotin, P. & Falk, H. L. (1958) *Cancer (Philad.)*, **11**, 473

Kotin, P., Falk, H. L. & Thomas, M. (1956) *Cancer (Philad.)*, **9**, 905

Kulp, J. L., Eckelmann, W. R. & Schulert, A. R. (1957) *Science*, **125**, 219

Largent, E. J. (1952) *The effects of air-borne fluorides on livestock.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 64

McCabe, L. C. & Clayton, G. D. (1952) *A.M.A. Arch. industr. Hyg.*, **6**, 199

Machle, W., Thamann. F., Kitzmiller, K. & Cholak, J. (1934) *J. industr. Hyg.*, **16**, 129

Madsen, L. C. (1942) *Keeping livestock healthy.* In: United States Department of Agriculture, *Yearbook, 1942*, Washington

National Academy of Sciences, National Research Council (1956a) *Publication 452*, Washington

National Academy of Sciences, National Research Council (1956b) *The biological effects of radiation ; a report to the public*, Washington

Pattle, R. E. & Cullumbine, H. (1956) *Brit. med. J.*, **2**, 913

Phillips, P. H. (1952) *Fluorosis in livestock.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 123

Publ. Hlth Bull. (Wash.), 1949, No. 306

Russell, E. S. (1955) *Brit. med. J.*, **1**, 826

Stokinger, H. E. (1954) *A.M.A. Arch. industr. Hyg.*, **9**, 366

Stokinger, H. E., Wagner, W. D. & Wright, P. G. (1956) *A.M.A. Arch. industr. Hlth*, **14**, 158

Van Middlesworth, L. (1956) *Science*, **123**, 982

United States Department of the Army (1957) *Department of the Army Pamphlet No. 39-3*, Washington

United States Department of Health, Education, and Welfare, Public Health Service (1956) *Proceedings of the Air Pollution Research Planning Seminar*, Washington

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EFFECTS OF AIR POLLUTION ON PLANTS

There are three principal air pollutants of major interest to agriculture—viz., sulfur dioxide, fluorine compounds, and smog. The last is a complex mixture, only partially understood at this time. There are at least two distinct types of smog, with many intermediate grades: the London type, which is a mixture of coal smoke and fog with enough sulfur dioxide to impart reducing properties to the mixture; and the highly oxidizing Los Angeles type, which usually contains neither coal smoke nor fog, but rather is a mixture of ozone and peroxidized organic compounds formed by photochemical reactions between oxides of nitrogen and innocuous organic compounds such as gasoline vapours or partially burned fuel. In addition to the two types of smog, certain organic compounds, such as ethylene, DDT, and some heterocyclic bases, are known to have powerful phytotoxicity and have done considerable plant damage in some locations. The subject has been reviewed in three recent publications (Thomas, 1951; Thomas & Hendricks, 1956; Zimmerman, 1955).

Sulfur Dioxide [1]

Sulfur dioxide has been studied longer and more intensively than the other pollutants. The older German studies are reported in several books (Haselhoff, Bredemann & Haselhoff, 1932; Haselhoff & Lindau, 1903; Stoklasa, 1923; Wislicenus, 1914), and in America there is an extensive literature (Dean & Swain, 1944; Holmes, Franklin & Gould, 1915; Katz, 1949, 1952; National Research Council of Canada, 1939; O'Gara, 1922; Setterstrom & Zimmerman, 1939; Setterstrom, Zimmerman & Crocker, 1938; Thomas et al., 1943, 1944; Thomas, Hendricks & Hill, 1944, 1950a, b, 1952; Thomas & Hill, 1935, 1937; Zimmerman & Crocker, 1934a,b).

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[1] For illustrations of typical lesions on a number of plants caused by sulfur dioxide, see Plate 1 (facing page 240).

The effects of sulfur dioxide on plants are fairly well understood. The gas is absorbed into the mesophyll of the leaves through the stomata. Toxicity is due largely to the reducing properties of the gas. The limiting concentration that can be tolerated in the cells is about the same for many diverse species, including water plants. When this concentration is exceeded, the cells are first inactivated with or without plasmolysis, then killed. When extensive areas are killed, the tissues collapse and dry up, leaving a characteristic pattern of interveinal and marginal acute injury. If only a few cells in an area are injured, this area may become chlorotic or brownish red in colour, owing to chronic injury. A slow oxidation of sulfite to sulfate occurs in the cells. This reduces the toxicity of the sulfite by a factor of about 30. Consequently, if the sulfur dioxide is not added to the system too rapidly, a rather large amount may be added without causing injury before sulfate toxicity occurs. Sulfate toxicity is a form of chronic injury manifested by white or brownish-red turgid areas on the leaf caused by the rupture of some cells or of chloroplasts within the cells (Solberg & Adams, 1956; Thomas & Hendricks, 1956; Thomas & Hendricks, unpublished data). Abscission often occurs at or before this stage. Photographs of typical sulfur dioxide injury are given in reports by the National Research Council of Canada (1939) and Thomas & Hendricks (1956).

Injury by sulfur dioxide is local. No systemic effects have been observed. While the injured areas of the leaves never recover, the uninjured areas quickly and fully regain their functions and new leaves develop normally.

It should be noted that there are many lesions caused by diseases and environmental conditions which resemble more or less those due to sulfur dioxide but have no connexion with the gas. These include white spot in alfalfa, frost or winter injury, tip burn, chlorosis, salt injury in wheat and barley due to uptake of too much salt from very slightly saline soil when growth is slow, and nutritional or senescence effects often seen in corn. Massey (1952) has discussed the problem of sulfur dioxide and hydrogen fluoride injury in relation to other " disease " symptoms, pointing out many of the difficulties encountered in making a definite diagnosis of gas injury. Evidently there are many situations in which it is impossible to diagnose gas damage with certainty by symptoms alone.

" Invisible " injury was postulated by Stoklasa (1923) as interference with the growth or functioning of the plant without attendant lesions on the leaves or elsewhere. A number of sulfur dioxide fumigation studies have been made for the purpose of investigating this theory (Hill & Thomas, 1933; Katz, 1949; National Research Council of Canada, 1939; Setterstrom et al., 1938; Swain & Johnson, 1936). No significant reduction in yield of the crops was found if the area of visible markings on the leaves was less than about 5 %. Measurements of photosynthesis (National Research Council of Canada, 1939; Thomas & Hill, 1937) also indicated that fumigations which did not cause leaf injury did not reduce carbon assimilation by a significant amount, though a temporary lowering of the photosynthesis was observed. Possibly a fumigation treatment could be devised that would produce a significant amount of invisible injury, but it would probably be of academic interest only. The details of the photosynthesis studies are given later. The effects of coal smoke in Manchester, discussed later (Bleasdale, 1952b), could constitute invisible injury due at least in part to sulfur dioxide, but this has not yet been definitely established.

Sulfur is an essential element in plants and is present in many biologically active compounds such as methionine, cystine, glutathione, thioctic acid, coenzyme A, thiamine, and many others. The organic sulfur fraction in the leaves of different plants is fairly constant in amount (Thomas et al., 1950b). The needles of conifers usually have about 0.1 % S dry basis (Katz, 1952); many broad-leaved plants have 0.15 %-0.3 %; and plants such as the crucifers may contain 0.6 %. Sulfate may be present in the leaves in a wide range of concentrations, without appreciably affecting the organic sulfur level. When sulfur dioxide is absorbed into the leaves in sublethal amounts, it is primarily oxidized to sulfate (Thomas et al., 1943; Thomas, Hendricks & Hill, 1944) but some may be reduced to organic sulfide. It has been shown by tracer (^{35}S) experiments (Steward et al., 1951; Thomas et al., 1944; Thomas, Hendricks & Hill, 1950b) that the final disposition is the same whether the element is absorbed through the roots as sulfate or through the leaves as sulfur dioxide—though sulfate is more effective as a nutrient than is sulfur dioxide, because of its greater mobility in the plant. An explanation is thus afforded for the fact that sulfur dioxide is local in its action as a toxicant and does not produce systemic effects.

Many suggestions have been made as to the mechanism of sulfur dioxide injury. Haselhoff & Lindau (1903) considered that the gas reacted with the sugars in the leaves, and was slowly released later with resultant injury to the cells. This suggestion seems improbable in view of the fact that the leaves are more susceptible to sulfur dioxide in the morning, when the sugars are low, than in the afternoon, when they are higher. Noack (1929) suggested that the sulfur dioxide reacts with the iron in the chloroplasts, thus interfering with its catalytic properties. Secondary photo-oxidative processes are then promoted which cause decomposition of the chlorophyll and death of the cells. Dorries (1932) postulated phaeophytin formation due to acidification of the chlorophyll by the sulfur dioxide. Experimental evidence on this reaction in the leaves of different species is conflicting. Another suggestion, made by Bleasdale (1952b) and based on the work of Hammett (1930), postulates an equilibrium between sulfhydryl groups and more-oxidized sulfur compounds, particularly sulfites. The former, which promote cell division and growth, are increased in amount but are inactivated by the latter. When the sulfite level is reduced, the level of sulfhydryl activity is increased. Presumably if the sulfite level is raised too much, permanent injury to the cells would result.

It seems clear that the toxicity of sulfur dioxide is caused primarily by its oxidation-reduction properties rather than by its acidity. Sulfur dioxide is considerably more toxic than hydrogen chloride, whereas it yields a weaker acid. Sulfite toxicity appears to be about 30 times the equivalent sulfate toxicity, as already noted.

Relative susceptibility of species

The different species of plants vary over a considerable range in their susceptibility to injury by sulfur dioxide. These differences seem to be due

TABLE 1. RELATIVE SENSITIVITY OF CULTIVATED AND NATIVE PLANTS TO INJURY BY SULFUR DIOXIDE (DETERMINED BY O'GARA)

Sensitive		Intermediate		Resistant	
CULTIVATED PLANTS					
Alfalfa	1.0[a]	Cauliflower	1.6[a]	Gladiolus (1.1-4.0)[b]	2.6[a]
Barley	1.0	Parsley	1.6	Horse-radish	2.6
Endive	1.0	Sugar beet	1.6	Sweet cherry	2.6
Cotton	1.0	Sweet William	1.6	Canna	2.6
Four o'clock	1.1	Aster	1.6	Rose	2.8-4.3
Cosmos	1.1	Tomato (1.3-1.7)[b]	1.7	Potato (Irish)	3.0
Rhubarb	1.1	Eggplant	1.7	Castor bean	3.2
Sweet pea	1.1	Parsnip	1.7	Maple	3.3
Radish	1.2	Apple	1.8	Boxelder	3.3
Verbena	1.2	Catalpa	1.9	Wisteria	3.3
Lettuce	1.2	Cabbage	2.0	Mock orange	3.5
Sweet potato	1.2	Hollyhock	2.1	Honeysuckle	3.5
Spinach	1.2	Peas	2.1	Hibiscus	3.7
Bean	1.1-1.5	Gooseberry	2.1	Virginia creeper	3.8
Broccoli	1.3	Zinnia (1.2)[b]	2.1	Onion	3.8
Brussels sprouts	1.3	Marigold	2.1	Lilac	4.0
Pumpkin	1.3	Hydrangea	2.2	Corn	4.0
Table beet	1.3	Leek	2.2	Cucumber	4.2
Oats	1.3	Begonia	2.2	Gourd	5.2
Bachelor's-button	1.4	Rye (1.0)[b]	2.3	Chrysanthemum	5.3-7.3
Clover	1.4	Grape	2.2-3.0	Snowball	5.8
Squash (1.1-1.4)[b]	1.4	Linden	2.3	Celery	6.4
Carrot	1.5	Peach	2.3	Citrus	6.5-6.9
Swiss chard	1.5	Apricot	2.3	Cantaloupe (muskmelon)	7.7
Turnip	1.5	Kale	2.3	Arbor vitae	7.8
Wheat	1.5	Nasturtium	2.3	Currant blossom	12.0
		Elm	2.4	Live oak	14.0
		Birch	2.4	Privet	15.0
		Iris	2.4	Corn silks and tassels	21.0
		Plum	2.5	Apple blossoms	25.0
		Poplar	2.5	Apple buds	87.0

TABLE 1 *(continued)*

RELATIVE SENSITIVITY OF CULTIVATED AND NATIVE PLANTS TO INJURY BY SULFUR DIOXIDE (DETERMINED BY O'GARA)

Sensitive		Intermediate		Resistant	
		NATIVE PLANTS			
Gaura (1.0)[b]		Dandelion	1.6	Purslane	2.6
Tobacco tree (*N. glauca)*	1.0	Orchard grass	1.6	Sumac	2.8
June grass (*B. tectorum)*	1.0	Rough pigweed (Redroot)	1.7	Shepherd's purse	3.0
Prickly lettuce	1.0	Black mustard	1.7	Milkweed	4.6
Mallow	1.1	Smartweed	1.8	Salt grass	4.6+
Ragweed	1.1-1.2	Lamb's quarters	1.8	Pine [c]	7-15.0
Curly dock	1.2	Sweet clover	1.9		
Buckwheat	1.2-1.3	Nightshade	2.0		
Bouncing bet	1.3	Hedge mustard	2.1		
Plantain	1.3	Cocklebur	2.3		
Sunflower	1.3-1.4	Tumbling mustard	2.4		
Rye grass	1.4				

Source: Thomas & Hendricks (1956). (By permission from "Air pollution handbook", by P. L. Magill, R. F. Holden & C. Ackley, editors. Copyright 1956. McGraw-Hill Book Company, Inc.)

[a] Factors of relative resistance compared with alfalfa as unity.

[b] More probable factors based on later experience.

[c] Data for pine represent October fumigations in Palo Alto, Calif., of Monterey, white, Allepo, and Coulter varieties. O'Gara factors calculated from the data of Katz & McCallum (National Research Council of Canada, 1939) are as follows: larch in May, 1.5; Douglas fir in May, 2.3; yellow pine (year-old) seedlings in May, 1.6, in August, 2.4-4.7.

primarily to differences in the rate of absorption of the gas by their leaves. Plants with succulent leaves of high physiological activity are generally sensitive. Examples are alfalfa, the grains, squash, cotton, grapes, and many similar plants. An exception is corn, which keeps its stomata tightly closed a great deal of the time. Plants with fleshy leaves and needles are inclined to be resistant, except when newly formed before cutinization. Examples are citrus, pine needles, and privet.

Table 1 gives factors of relative sensitivity to sulfur dioxide, found by O'Gara (cited by Thomas & Hendricks, 1956) for about 100 plants based on alfalfa as unity. The factors were obtained by fumigating each plant for an hour with different concentrations of the gas until the concentration is found that causes traces of injury. The relative humidity is also measured and the concentration is adjusted to 100% relative humidity by a relationship that is given later. This value is divided by 1.25 since alfalfa requires about 1.25 p.p.m. for incipient injury in one hour. In a few cases more probable values are noted, based on later experience. Great variability is noted in the susceptibility of the varieties of some species, such as the conifers, gladiolus, and rose. Others, such as prunus, have more constant factors.

For these reasons, the O'Gara factors are suggestive only, and are subject to variation with different climates, varieties, and stages of growth.

Environmental factors

The same conditions that enhance the absorption of the gas predispose the plant to injury. They include high light intensity (especially in the morning hours), high relative humidity, adequate moisture supply, and moderate temperatures. These are the conditions that cause the stomata to open. Loftfield (1921) demonstrated the existence of a relationship between the diurnal trend of stomatal apertures and susceptibility in alfalfa and other plants. This was confirmed for alfalfa by Katz & Ledingham (National Research Council of Canada, 1939). Most plants close their stomata at night and are therefore much more resistant in the dark than in the light. Those that do not, such as the potato, are about equally sensitive under these conditions. Individual alfalfa leaves, observed to be open at night, were found to be as sensitive as in the day.

TABLE 2

EFFECT OF RELATIVE HUMIDITY ON ALFALFA AND OTHER PLANTS

Relative humidity, %	Relative sensitivity	Relative resistance
100	1.00	1.00
80	0.89	1.12
60	0.77	1.30
50	0.69	1.45
40	0.54	1.85
30	0.31	3.2
20	0.18	5.5
10	0.13	7.7
0	0.10	10.0

Source: Thomas & Hendricks (1956). (By permission from "Air pollution handbook", by P. L. Magill, F. R. Holden & C. Ackley, editors. Copyright 1956. McGraw-Hill Book Company, Inc.)

The stomata also close under conditions of moisture stress—which explains the observed relationship between moisture and susceptibility. Table 2 gives the relative susceptibility or resistance of alfalfa at different values of the relative humidity. The range of these factors shows the necessity of considering the relative humidity in connexion with susceptibility observations.

It has been found that the susceptibility falls off in the afternoon of a cloudless day while the light intensity is still high and all the other conditions remain constant. It is probable that the build-up of carbohydrates in the leaf serves to reduce the susceptibility to some extent, possibly by aldehyde-sulfite reactions.

Exposure equations

As a result of a great many brief fumigations of alfalfa, O'Gara (1922) concluded that a reciprocal relation existed between exposure time (t = hours) and concentration (C = p.p.m.) which causes traces of injury under conditions of maximum sensitivity, as follows:

$$(C - 0.33)t = 0.92 \qquad (1)$$

The constant 0.33 p.p.m. represents a concentration that presumably can be endured indefinitely. This, of course, is not true of prolonged fumigations which encompass conditions not encountered in the short exposures. In one hour, $C = 1.25$ p.p.m.

Thomas & Hill (1935) generalized this equation for any degree of leaf destruction and any degree of susceptibility. With maximum susceptibility, three equations were calculated as follows:

$(C - 0.24)t = 0.94$	Traces of leaf destruction	(2)
$(C - 1.4)\ t = 2.1$	50 % leaf destruction	(3)
$(C - 2.6)\ t = 3.2$	100 % leaf destruction	(4)

The work also indicated that the toxic dose of sulfur dioxide to the leaves, if present as unoxidized sulfite, would be about 1350 p.p.m. S in the dry tissue. If the active cell substance is about half the total leaf substance, this value would be about 2700 p.p.m. in the cells.

Effects of sulfur dioxide on photosynthesis and respiration

The photosynthesis and respiration of plants can be measured automatically and without interfering with their growth beyond confining them in a chamber through which a measured stream of air is passed. To this system the fumigant can be added, at will, in any desired concentration. Illumination with either sunlight or artificial light may be applied, and control of temperature and relative humidity is practicable. The carbon dioxide content of the air is then determined as it enters and leaves the chamber. The difference represents carbon dioxide, assimilated or respired, and effects due to the fumigant are immediately evident. Several arrangements of the fumigation equipment have been described (National Research Council of Canada, 1939; Thomas & Hill, 1937), and detailed graphs of the carbon dioxide exchange are illustrated in Fig. 4 (page 261). It is practicable to continue treatments for weeks or for months and to measure the gas exchange day and night during this time.

Thomas & Hill (1937) made a detailed study of the action of sulfur dioxide on alfalfa, using this method. It was shown that when a high concentration of gas was employed for a period too short to cause appreciable leaf lesions, the rate of photosynthesis fell sharply to a lower level, then recovered and reached its normal level in a few hours or next day. With lower concentrations—for example, 0.4 or 0.6 p.p.m. for four hours—definite reduction in assimilation occurred, with complete recovery within about two or three hours after discontinuing the fumigation.

At still lower concentrations there was no direct interference by the gas with assimilation except in some continuous fumigations. Brief summaries of these experiments follow.

(1) About 0.3 p.p.m. applied four hours per day for a month had no effect, though the sulfur content of leaves reached 1.7 %.

(2) Continuous application of 0.24 p.p.m. intake concentration (0.18 p.p.m. average of intake and outlet) reduced assimilation by 1 % during the first three days and by 3 % during the next three days. From the 17th to the 22nd day the reduction was 24 %; during three days following the termination of the fumigation, it was 19 %. The leaves had 2 % acute markings and were somewhat chlorotic, having low chlorophyll values. They contained 2 % sulfur at this time, which represents nearly the maximum before abscission. Dead tissue on the treated plot represented 22 % of the total as compared with 12 % on the check plot. Senescence and sulfate toxicity can therefore explain most of the effects on assimilation. The final 5 % recovery of assimilation probably represents the " invisible injury " level of the fumigation after about the first six days.

(3) Similar results followed continuous treatment with 0.19 p.p.m. intake (0.14 p.p.m. average of intake and outlet), except that it required 11 days before any reduction of assimilation was noted, which finally amounted to about 10 % after an exposure of 26 days. At that time the leaves carried 1.35 % sulfur. After stopping the fumigation, the assimilation level rose to 95 % of the control. Only traces of leaf injury were apparent, but the chlorophyll content of leaves was depressed slightly. Some abscission of older leaves had also occurred. When the fumigation was resumed at 0.29 p.p.m. intake (0.21 p.p.m. average of intake and outlet) for 14 days, results fully confirming those of the preceding experiment were obtained.

(4) Finally, a 45-day fumigation with 0.14 p.p.m. intake (0.10 p.p.m. average of intake and outlet) showed no significant effect on assimilation or growth of the alfalfa. Concentration of sulfur in the leaves reached 1.5 %. The chlorophyll content was not affected. There was some abscission of the older leaves, but new growth was sufficient to maintain the activity of the plants. Increase of assimilation after stopping the fumigation was not more than 2 %, indicating that the sulfite level in the cells was probably very low.

PLATE 1

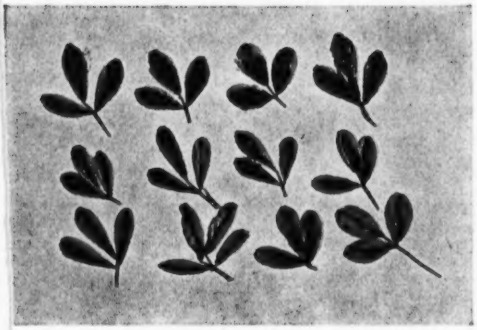

Fig. 1. Acute SO_2 damage on alfalfa

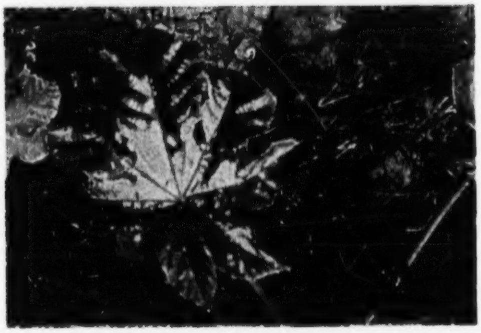

Fig. 2. Acute SO_2 damage on castor bean

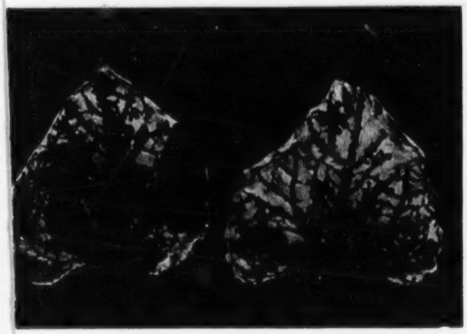

Fig. 3. Acute SO_2 damage on pinto bean

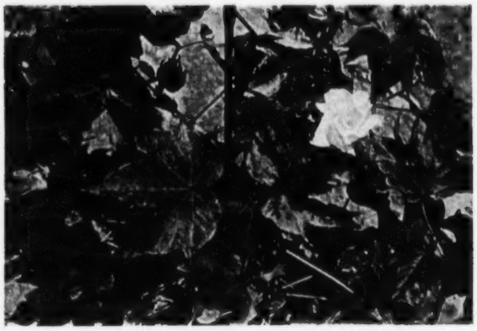

Fig. 4. Chronic SO_2 damage on cotton

Acknowledgement is made on page 275 to the persons who furnished the above coloured photographs.

Katz et al. (National Research Council of Canada, 1939) carried out some similar experiments and obtained similar results.

Leaf destruction in relation to yield

Many industries that emit phytotoxic gases are located near valuable agricultural land. If damage is done to nearby crops, the farmer is legally entitled to recover his losses. Therefore an objective method is required for determining the extent of the losses. Many studies have shown that, for sulfur dioxide and a number of field crops, the reduction in yield of a crop is proportional to the percentage of leaf area destroyed:

$$y = a - bx \tag{5}$$

where y is the yield expressed as the percentage of full yield in the absence of the pollutant, and x is the percentage of leaf area destroyed estimated on the leaves present at the time of fumigation. The constant, a, is about 100 %; b is the slope of the yield/leaf-destruction curve.

Alfalfa

Hill & Thomas (1933) fumigated alfalfa plots with sulfur dioxide once, twice or three times during the growth of the crop, allowing 10-14 days between successive treatments and between the final treatment and the harvest of the crop. The percentage of leaf destruction, x, was determined by counting the marked leaves on representative samples of the plot and estimating the average area of the marked leaves that was injured. Each treatment of the multiple fumigations was measured separately. The yield, y, of each fumigated plot was expressed as a percentage of the average yield of several adjacent control plots.

The equations obtained were as follows:

(*a*) Single fumigation early, medium, or late in the growth of the crop:

$$y = 99.5 - 0.30x \qquad n = 96 \qquad r = 0.64 \pm 0.06 \qquad Sy = 7.4\,\% \tag{6}$$

(*b*) Two fumigations on the crop:

$$y = 95.5 - 0.49x \qquad n = 34 \qquad r = 0.79 \pm 0.07 \qquad Sy = 8.2\,\% \tag{7}$$

(*c*) Three fumigations on the crop:

$$y = 96.6 - 0.75x \qquad n = 12 \qquad r = 0.98 \pm 0.01 \qquad Sy = 4.1\,\% \tag{8}$$

In other experiments, approximately the same relationships were found by picking off the leaves instead of destroying them with sulfur dioxide, suggesting that the action of the latter is limited to its visible effects.

The effects of multiple fumigations are additive. A given percentage of leaf destruction reduces the yield by the same amount regardless of stage of growth, even though, for example, three times as much leaf tissue is destroyed when the crop is three-quarters grown as when one-quarter grown. It is assumed that the fumigated and control crops have the same growth periods. If the fumigations are so severe that the reserves of carbohydrates in the roots are not adequately replenished after new leaves are developed to replace those destroyed, growth of the next crop may be curtailed. A minimum leaf destruction of about 50 % at least a week before harvest is needed before this effect becomes measurable on the subsequent crop.

The alfalfa yield data of Katz & Ledingham (National Research Council of Canada, 1939) give an equation that is not appreciably different from equation (6).

$$y = 99 - 0.37x \qquad n = 103 \qquad r = 0.48 \qquad Sy = 8.8\,\% \tag{9}$$

The fumigation treatments in equation (9) ranged from 1 to 600 hours and from 0.1 to 3 p.p.m., whereas most of the treatments for equations (6) to (8) were of only 1-2 hours' duration and 1-5 p.p.m.

Wheat, barley and cotton

The grains differ from alfalfa in that vegetative growth is not the primary consideration but, rather, production of kernels. The stage of development of the plant when the leaf destruction occurs becomes very important. During tillering, severe leaf destruction may be sustained without appreciable effect on yield. The plant simply develops more tillers and/or enlarges the injured leaves so that its ultimate growth is not interfered with. After the shoots form, the effect of leaf destruction becomes increasingly important. It is greatest in the blossom stage, but only a little less important in the boot and milk stages. In the dough stage when the plant is maturing, the effect is greatly reduced. The plant is beginning to dry up at this time.

Equations for barley, calculated from the data of the Selby Smelter Commission (Holmes et al., 1915), are as follows:

Early stage, less than 25 cm in height:

$$y = 98 - 0.06x \qquad n = 18 \qquad r = 0.13 \qquad Sy = 12.2\,\% \tag{10}$$

This equation lacks significance because of its small slope.

Heading-out stage and later (some plots fumigated earlier also, as well as throughout the ripening period):

$$y = 98 - 0.40x \qquad n = 60 \qquad r = 0.74 \qquad Sy = 10.2\,\% \tag{11}$$

An equation for winter wheat under average field conditions, calculated from data by the American Smelting and Refining Company, is the following:

$$y = 103 - 0.37x \qquad n = 71 \qquad r = 0.75 \qquad Sy = 10.2\,\% \tag{12}$$

Brisley & Jones (1950) studied Sonora wheat in Arizona from 1941 to 1944. They fumigated 10 plots each week for 14 weeks, attempting to

TABLE 3

SLOPE (b) OF THE REGRESSION CURVE OF WHEAT, $y = a - bx$, AS THE PLANT DEVELOPS

Weeks from planting	Functional leaf area, % of total	b
1-7	0.1-11	0.01
7	11	0.08
8	22	0.16
9	37	0.28
9-14	37-67	0.35
11-14	50-67	0.37

Source: Brisley & Jones (1950).

produce a wide range of leaf destruction in each group of plots. They measured the leaf area destroyed and expressed the result as a percentage of the total amount of leaf area produced by the plant during its whole growth period. There was never more than 67 % of active tissue present at any one time and, in the last week, the functional tissue fell to 46 % of the total. The 1944 data gave the equation:

$$y = 100 - 0.66z \qquad n = 130 \qquad r = 0.95 \qquad Sy = 2.2\,\% \qquad (13)$$

where z represents leaf destruction calculated on the basis of total leaf area, to distinguish it from x, calculated on the leaf area at the time of fumigation. Using this equation, the relationship between y and z is independent of the stage of growth of the plant. This would seem to be a definite advantage. However, it must be remembered that the determination of the total leaf area produced by fully matured control plants is a task involving some uncertainty as well as considerable labour.

When the data for the various weeks (Table 3) are calculated separately, using x instead of z, the values of the constant, b, increase with the age of the plant and approach closely the values in equations (11) and (12). The method of calculation based on z and the method based on x in a series of equations like those in Table 3 should give equally satisfactory results.

The grain data so far considered represent plants grown under average conditions of fertility and moisture supply. With greater fertility or water supply, the effect of leaf destruction in reducing the yield is lessened, so that in an extreme case of a field yielding over 60 bushels of wheat per acre (150 bushels per hectare), even very severe injury by sulfur dioxide did not cause appreciable reduction in yield (Thomas & Hendricks, 1956). Data by Brisley & Jones (1950) during more favourable crop years than the one considered indicated much smaller values of the regression coefficients than those given. Evidently local and seasonal conditions need consideration in the application of these equations.

Brisley, Davis & Booth (1959) have published an equation for cotton that is almost identical with their equation for wheat. The interpretation is similar to that given for wheat.

$$y = 99 - 0.68z \qquad n = 494 \qquad r = 0.84 \qquad Sy = 5.5\% \qquad (14)$$

Data on other crops have not been published, but a brief reference may be made to some of the work that has been done in order to indicate its scope. Each crop presents special problems. For example, yields of tomatoes and cotton vary according to the length of the harvest season and would be particularly affected by early frost. The question of including green tomatoes in the yield must be considered. Cotton develops a great deal of chronic injury, which appears to have a smaller effect in reducing yield than acute injury, necessitating a decision as to weighting the two kinds of markings. Large fumigation plots have advantages over small plots for plants of indeterminate and variable size such as tomatoes and cotton, but large plots require special fumigation equipment.

Mid-season fumigations of these crops, when flowering and fruit formation were most active, gave reductions in yield ranging from the values found for alfalfa and grain to nearly double those values under various conditions. The reductions were small in earlier fumigations, as with wheat and barley. Evidently the responses of all these crops to leaf destruction by sulfur dioxide are similar. The responses may vary over a considerable range in different seasons and in different environments, particularly as regards soil fertility and available moisture.

Fluorine Compounds[1]

Fluorides in general, and gaseous fluorides in particular, have assumed great importance as air pollutants during the past decade, first, because hydrogen fluoride and silicon tetrafluoride are toxic to some plants in concentrations as low as 0.1 parts per thousand million (p.p.t.m.) and, secondly, because all fluorides, particulate as well as gaseous, may be accumulated by forage to build up concentrations in excess of 30-50 p.p.m. on the inside and outside of the leaves. No lesions are ordinarily observed in the forage. This vegetation, when consumed by cattle or sheep, can cause fluorosis in the animals. This phase of the problem will not be considered further in this chapter.[2] However, it often has even greater economic significance than the direct plant-damaging effects of fluorides.

The toxic effects of fluorides on vegetation present many paradoxes. Some plants are injured by extremely low concentrations in the atmosphere and/or in the tissue; others can withstand more than a hundredfold as much. The reason for this enormous range of tolerance is not apparent. A few factors may be mentioned which perhaps play a part.

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[1] For illustrations of typical lesions on a number of plants caused by hydrogen fluoride, see Plate 2 (facing page 248).

[2] See the chapter "Effects of Air Pollution on Animals," by E. J. Catcott (section on animal fluorosis, page 227).

(1) Fluorides absorbed in the leaves are translocated towards the margins of broad leaves or the tips of leaf blades. By this means the body of the leaf, where absorption takes place, may remain at a low level of concentration while the margins build up lethal concentrations, causing marginal necrosis. Gradients of 2 or 3 to 1 up to 100 to 1 have been observed in the tips or margins as compared with the body of the leaves. Appreciable translocation of fluoride from the leaf to other parts of the plant does not occur.

(2) Insoluble fluoride salts may be precipitated in the tissue, reducing the activity of the element. This seems to occur to some extent in plants of high ash-content (Thomas & Hendricks, unpublished data), but different varieties of gladiolus, varying considerably in susceptibility to hydrogen fluoride, have been found to have identical ash contents.

(3) Part of the hydrogen fluoride may be adsorbed on the exterior of the leaf. This can be washed off with water. It represents a variable but substantial part of the total fluoride absorbed. Some may be lost by volatilization.

(4) Organic fluorine compounds may be formed in the leaves. There is some evidence from partition studies in organic solvents that such compounds are formed, but definite substances have not been isolated, and their significance is unknown (Thomas & Hendricks, unpublished data; Zimmerman & Hitchcock, 1956). It is well known that fluoracetate occurs in the poisonous African plant, *gifblaar*. It has been found to block the citric acid cycle by forming monofluorcitrate (Peters, Wakelin & Buffa, 1953).

(5) Fluorides can inhibit certain plant enzymes in very low concentrations (Warburg & Christian, 1941). For example, plant enolase in vitro is measurably inhibited by 3 p.p.m. fluoride. McCombie & Saunders (1946) state that cholinesterase is inhibited by 10^{-10}M di-isopropyl fluorophosphonate, whereas di-ammonium fluorophosphonate requires 10^{-2}M to cause inhibition. Whether the latter fact has any bearing on fluorine toxicity in plants is not known, but it illustrates a large difference in biological effect due to a relatively small difference in structure.

Sources of fluorides

The industries mainly responsible for emitting fluorides into the atmosphere are (*a*) aluminium reduction, (*b*) smelting of iron and non-ferrous ores, (*c*) ceramics, and (*d*) phosphate reduction and phosphate fertilizers. The fluorides come principally by volatilization from the molten cryolite bath in the aluminium industry, and from impurities in the raw materials in the others. Hydrogen fluoride and silicon tetrafluoride are released by high-temperature reactions involving water, silica and fluoride, and by treating with acid the fluorides always present in phosphate rock. Reduction of phosphate rock liberates the fluorides as in the other high-temperature processes.

Chronic fluoride problems have doubtless been present from the beginning of the aluminium and fertilizer industries, though there has been serious litigation in North America only in recent years. In Europe, fluoride injury was recognized 50 years ago (Bredemann & Radeloff, 1937a, b; Haselhoff et al., 1932; Radeloff, 1938). The non-ferrous smelting industry has escaped complaints owing to the small fluoride output that is discharged through tall stacks. In general, the ceramic industry involves small units with limited fluoride output, which affects only the immediate locality.

Remedial measures have been taken by nearly all the larger fluoride-emitting industries, in some cases at great cost. Most of the aluminium and fertilizer plants scrub their waste gases with water to remove at least 85 %-90 % of the gaseous fluoride. One aluminium plant has built a great stack 500 feet (about 150 m) high through which to discharge its gases, with forced draught and after scrubbing as necessary. Thus, many serious problems have been solved or minimized. Others remain. For example, the ceramic industry would be hard-pressed if required to process its stack gases for the removal of the small amount of fluoride emitted. Further, coal may contain from 85 to 295 p.p.m. fluoride (Churchill, Rowley & Martin, 1948), which is largely emitted on combustion and adds to the contamination of urban atmospheres.

Fluorides in the air

Though fluoride is generally present in soils to the extent of several hundred parts per million, plants show little tendency to take it up, first, because of its insolubility and, secondly, because the fine roots have an effective mechanism for excluding it, especially if the pH of the soil is high. Sodium fluoride is taken up rather readily but, of course, it is rarely present. Uptake from the soil has been studied by McIntire et al. (1949) and by Daines and his associates (Brennan, Leone & Daines, 1950; Daines, Leone & Brennan, 1952; Leone et al., 1948; Prince et al., 1949). Consequently, when a plant contains more than a few parts per million fluoride, atmospheric contamination is indicated. An exception is camelia, which took up 1500-2000 p.p.m. while peach took up 20 p.p.m. (Zimmerman & Hitchcock, 1956).

Large industrial sources of fluoride may release a few hundred to a few thousand pounds of gas and solids per day into the air. This may be compared with large industrial emissions of sulfur dioxide, which have ranged from about 50 to over 1000 tons (calculated as elemental sulfur) per day. Consequently, whereas ground concentrations of sulfur dioxide are often in the parts per million range, those of fluoride would be in parts per thousand million.

Only a limited number of air analyses for fluoride are found in the literature. Kehoe (1949) reported 3-6 p.p.t.m. in the atmosphere of Cincinnati. Largent (1949) found 0-29 p.p.t.m. near a superphosphate plant. At Fort William, Scotland (Agate et al., 1949), concentrations of 9-140 p.p.t.m. were found between 100 yards and 1 mile (90-1600 m) down-wind from an aluminium plant. In this area the vegetation contained 5-1000 p.p.m. Adams et al. (1952) made a systematic study near an aluminium plant near Spokane, Washington, in 1949-50. They found average eight-hour concentrations from 5 to 18 p.p.t.m. at 12 stations and maximum concentrations of 11-147 p.p.t.m. This area was re-sampled for over two weeks in July 1951 (Adams et al., 1956). Average concentrations at three locations ranged from 0.5 to 2.4 p.p.t.m., with a few four-hour maxima as high as 20 and 30 p.p.t.m. At that time this plant emitted its contaminants through louvers in the roof of the pot line buildings without any collection. A considerable area of ponderosa pine near the plant was killed (Miller et al., 1952). Of course, with the installation of scrubbing and collection devices, field concentrations have been greatly reduced, so that the foregoing values are much too high to be representative of current field concentrations adjacent to most of the large fluorine-emitting industries.

Fluorides in the field

Many field studies of the effects of fluorides on vegetation have been made. De Ong (1946) observed fluorine injury in a large orchard near an aluminium plant. Apricot, peach, prune, fig, and apple injury was described, including abscission of leaves and dropping of fruit. Miller, Johnson & Allmendinger (1948) analysed prune and gladiolus leaves from industrial areas and found 500-600 p.p.m. in the foliage. Miller et al. (1952) observed injury to ponderosa pine due to fluorides. The current year's needles (1949) had 129 p.p.m. while the 1946 needles carried 462 p.p.m. fluoride. McIntire et al. (1949) found up to 300 p.p.m. in the vegetation near aluminium and fertilizer plants in Tennessee.

In 1952 Kaudy et al. (1955) surveyed the fluorine content of citrus in Southern California with particular reference to a large steel plant at Fontana. In 1954, Brewer, McColloch & Sutherland (1957) extended the study to the commercial vineyards which were visibly injured by hydrogen fluoride in that locality. In both surveys, appreciable levels of fluoride were found in the citrus and grape leaves, being greatest near the plant in the principal down-wind directions and falling off with distance. The levels rose steadily as the growing season advanced. Maximum concentrations reached 211 p.p.m. (dry basis) in the spring cycle citrus leaves and 462 p.p.m. in the grape leaves. In the grape fruit the maximum was 2.7 p.p.m. (fresh weight basis). At 9 miles (14 km) to the north-east the levels reached 114 p.p.m. in grape leaves and 0.7 p.p.m. in the fruit. In other directions concentrations were less. Citrus leaves still showed 25 p.p.m. at a distance of 20 miles (32 km) to the west. In remote parts of the Los Angeles area, concentrations were about 1 p.p.m. in citrus, though in one heavily industrialized district as much as 57 p.p.m. were present from local sources.

Allmendinger, Miller & Johnson (1950) found that spraying the plants with lime gave rather effective protection from hydrogen fluoride fumigations, reducing the percentage of leaf destruction by a factor of 5 to 10. From 74 % to 94 % of the fluoride taken up could be washed from the surface of leaves, owing to its having reacted with the lime.

Description of lesions

Acute fluoride lesions on plants are quite characteristic, but are not the same in all species. The lesions vary also according to the gas concentration and length of exposure that produce them. High concentrations in the parts per million range with short period of exposures in the hour range produce lesions closely resembling those caused by sulfur dioxide. Such fumigations are unrealistic, however, since hydrogen fluoride concentrations in the field seldom exceed a few parts per thousand million. Lesions produced by prolonged exposures to low concentrations in the parts per thousand million or even the parts per ten thousand million range are of greatest interest. The following discussion is concerned with this type of fumigation.

Injury to gladiolus starts at the tip, and gradually extends down the blade as ivory-coloured necrotic tissue sometimes streaked with brown. A somewhat uniform front with a very narrow chlorotic band between the healthy and necrotic tissue is usually maintained. Islands of injured tissue surrounded by healthy tissue do not occur except with high fluoride concentrations. The pattern is similar on iris and the small grains. The current year's needles on pine, which are the most sensitive, are also similarly injured, except that the necrotic tissue is reddish brown. In apricot, prune, peach, grape, and other netted leaves, the injury is marginal with a sharp line of demarcation between healthy and necrotic tissue. At times the narrow transitional area is red. In the peach, the necrotic areas tend to become detached, leaving the leaf apparently healthy except for reduced size and a serrated edge. Abscission of the leaves of the fruit trees occurs readily even with moderate amounts of leaf destruction.

A different type of injury is produced on maize, consisting of a kind of mottled interveinal chlorosis, usually without marginal necrosis, since translocation of the element to the margins is very slow. It is often difficult to distinguish the fluoride symptoms from the discolorations due to incipient maturity.

PLATE 2

Fig. 5. Fluoride injury on grape

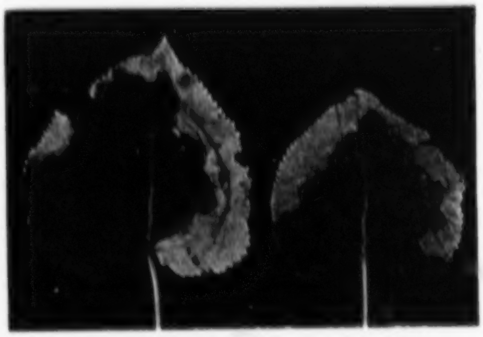


Fig. 6. Fluoride injury on apricot

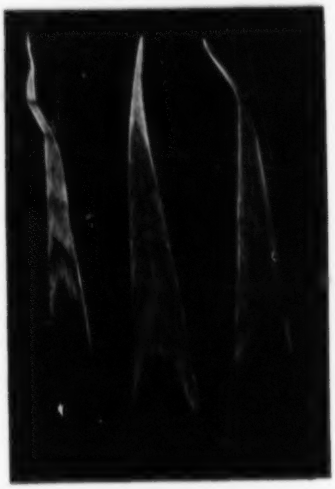


Fig. 7. Fluoride injury on Dutch iris

Fig. 8. Fluoride injury on loquat

Acknowledgement is made on page 275 to the persons who furnished the above coloured photographs.

In the dahlia, the very young leaves may be injured while emerging from their buds or while expanding rapidly. When this occurs they become almost black. More recently, similar lesions on young citrus and avocado leaves following exposure to about 5-10 p.p.t.m. hydrogen fluoride for several weeks were seen at Riverside, California. The result was nearly complete defoliation of the new shoots, whereas the older leaves were uninjured.

Large Washington navel orange trees, after 8-9 months' exposure to about 2 p.p.t.m. hydrogen fluoride, formed as many new shoots as the controls, but the shoots were less vigorous with shorter internodes and smaller leaves. Later these effects were hardly noticeable. Some of the young leaves showed tip necrosis and abscissed quickly. Interference with the development of new shoots thus appears to be a possible effect on many plants due to prolonged exposure to hydrogen fluoride in the p.p.t.m. range.

The petals of flowers are usually more resistant than the leaves. This is definitely true of gladiolus, though the petals can be injured by somewhat elevated concentrations. The petals of petunia are more easily injured than the leaves.

The European literature (Kotte, 1929; Radeloff, 1938) ascribed injury to the apical part of early plum and pear fruit to fluoride. Local softening of the tissue was involved. Injury to fruit due to atmospheric fluorides has not been reported in the USA other than by Griffin & Bayles (1952), who noted chocolate brown spots at the apical end of peaches together with split suture on trees that had been fumigated with an average concentration of 390 p.p.t.m. hydrogen fluoride (maximum 1460 p.p.t.m.) for 20.5 hours.

Anderson (1956) described the effects of fluoride sprays on peaches as follows:

> Sodium fluosilicate was experimented with for the control of bacterial spot of peach. While these sprays have been abandoned as peach sprays, the peculiar injury produced is of some interest. The condition brought about by fluoride sprays was called " soft suture " and resembles in many respects the virus disease of peach known as " red suture ". The fruit matures several days ahead of normal ripening. The suture takes on a yellowish-red to deep-red colour and softens even while the remainder of the fruit is quite firm. No decided difference in the taste of the fruit is noted, but the soft suture is easily bruised and the fruit becomes unsightly.

Benson (1959) has ascribed " soft suture " on peaches near Wenatchee, Washington, to gaseous fluoride or cryolite fumes from an aluminium plant. Extensive economic damage to the crops was noted. The fluoride content of the affected fruit peel was low (about 15-20 p.p.m.). The leaves had 44-112 p.p.m. Soft suture could be prevented by spraying the trees with lime, which evidently absorbed the fluoride on the leaf and fruit surfaces and also increased their calcium content.

Daines et al. (1952) noticed injury to peach foliage at some locations in New Jersey which was similar to that caused by fluoride sprays. Loss of crop due to poor set or early fruit drop was complained of by the growers.

Methods of analysis for fluoride

Definite diagnosis of fluoride exposure is readily accomplished by analysis of the air and vegetation. Methods of analysis are now available which are capable of giving reliable results. Some of the earlier work is probably unreliable. Nichol et al. (1957) discuss in detail field sampling of vegetation for fluoride analysis. These methods are particularly essential in forage sampling for the diagnosis of animal injury.

Fluoride in vegetation

In the method employed by the Stanford Research Institute (Remmert et al., 1953), a large representative sample of vegetation is dried and ground in a Wylie mill. In some laboratories the sample is treated with a known amount of fluorine-free lime before drying. A 5-g aliquot of the meal, containing lime or magnesia, is treated with water in an inconel dish, dried, and ashed at 600° C. The ash is fused with 5 g sodium hydroxide. The melt is dissolved in water and transferred to a distilling flask, from which the fluoride is steam-distilled at 135° C with perchloric acid or at 150° C with sulfuric acid (Smith & Parks, 1955), the final volume of the distillate being 500 ml.

A 200-ml aliquot of the distillate may be conveniently titrated with thorium nitrate (1 ml = 100 μg of fluoride) in a 250-ml Erlenmeyer flask. One ml of 0.05 % alizarin red solution is added to the flask. The colour is adjusted to neutrality by adding from burettes either 0.05 N perchloric acid or 0.05 N sodium hydroxide. Then 1 ml of a fresh 9.5 % monochloracetic acid solution containing 2 % sodium hydroxide is added to stabilize the pH at 3.0. The solution is titrated with thorium nitrate and compared with another flask containing distilled water similarly treated, to which 0.16 ml of the thorium solution has been added to give a faint pink colour. Colour changes due to 0.01 ml of titrant can be detected with a daylight fluorescent light and a table top of white glass and/or a large piece of filter paper.

In an alternative method extensively used in the author's former laboratory at the American Smelting and Refining Co., Salt Lake City, the large fresh sample is cut into small pieces with scissors, and a representative portion of about 20-30 g is homogenized in a Waring blendor with water and 20 ml of 5 % lime suspension. The slurry is transferred to a 250-ml wide-mouth screw-cap bottle for storage, with a drop of toluene added as preservative. For analysis the ground sample is made up to 300 or 500 ml volume, placed in a large beaker and stirred mechanically while aliquots are drawn out, using unconstricted pipettes of 50- and 80-ml capacity. One 50-ml portion is placed in a weighing dish for dry matter determination, and 50- and 80-ml portions are placed in 125-ml nickel crucibles for analysis. The latter are dried, then ashed at 600° C; the ash is fused with 5 g sodium hydroxide.

dissolved and steam-distilled as before. The remainder of the slurry is retained for additional analyses, if necessary.

The distillate may be titrated by adjusting a 200-ml aliquot to pH 3.3, using a pH meter and transferring to a 250-ml graduated cylinder covered on the outside with black matte paper and black insulating Scotch tape One ml of 0.032 % chrome azurol-S solution is added, and the solution is titrated against a distilled water blank in another cylinder, to which 0.02 ml of thorium nitrate is added after adjusting to pH 3.3. Illumination is from below over a fluorescent lamp in a box with a white glass cover. The solution is stirred during titration with a glass tube having a flat disk on the end. The end point is very sharp. In fact, small amounts of fluoride are generally titrated with thorium nitrate solution equivalent to 10 μg/ml for increased sensitivity.

Air samples are usually taken by aspirating distilled water at 1 cu.ft/min. (0.028 m^3) for periods up to 24 hours in large impinger bottles. Allowance for evaporation must be made and the volume of liquid should not be allowed to decrease below about 150 ml. The fluoride in these solutions may be concentrated and separated from any contamination by the ion exchange method of Nielsen & Dangerfield (1955) in preparation for the conventional thorium nitrate titration.

An application of the accumulating sulfur dioxide autometer has been described for the automatic collection of samples for fluoride (Thomas et al., 1952). The solutions having a volume of 100 ml are aspirated at 0.5 cu.ft/min. for three hours, then discharged into polyethylene bottles on a turntable. Direct titration of the solutions is usually possible, but interfering contamination may sometimes be present, and it is advisable to distil them. A sample can be added to the distillation flask containing perchloric acid, 200 ml steam-distilled out, then another sample added without changing the acid. Many more samples can be distilled from the same acid. With a battery of stills, a large number of distillations can be conveniently carried out in this way.

Fumigation experiments

Early fumigations with hydrogen fluoride were done at concentrations from about 0.05 to 2 p.p.m. (Daines et al., 1952; Griffin & Bayles, 1952; Zimmerman, 1949). Field conditions were not simulated. Compton & Remmert (personal communication, 1950) were the first to investigate the range below 14 p.p.t.m., producing characteristic lesions on gladiolus with these low concentrations. A five-week fumigation treatment at an average of 0.1 p.p.t.m. injured the tips of gladiolus for a distance of about 1 inch (2.5 cm) and increased the fluoride content of the tip to 148 p.p.m. for a length of 3 inches (7.5 cm). Appreciable injury developed in a few days with 1-2 p.p.t.m., and 4 inches (10 cm) of injury were produced by 10 p.p.t.m.

TABLE 4

RELATIVE SENSITIVITY OF CULTIVATED AND NATIVE PLANTS TO INJURY BY HYDROGEN FLUORIDE

Sensitive		Intermediate		Resistant	
CULTIVATED PLANTS					
Class 1		Class 3		Class 5	
Gladiolus	ABC	Corn (Golden Cross bantam)	B	Columbine and Canterbury bell	A
Pine (young needles)	B	Pepper (Calif. Wonder)	C	Dogwood and lilac	C
Apricot (Chinese)	BR	Raspberry (Washington)	C	Lobelia and petunia	A
Azalea	C	Aster and sweet William	A	Rose	B
Blueberry (Jersey)	C	Dahlia (immature leaves)	A	Apple (red Jonathan)	C
Prune (Italian PRH 1)	C	Dahlia	C	Live oak and pine	B
Tulip	B	Petunia petals	A	Tomato	ABC
Jerusalem cherry	B	Clover	B		
		Barley, flax, oats, and rye (young plants)	A	Class 6	
Class 2		Apple (Delicious), cherry (Bing)	C		
		Apple	B	Alfalfa (Ranger)	C
Milo maize	B	Birch, hawthorne, silver maple, mountain ash, mulberry, sycamore, and yellow willow	C	Corn (Golden Cross bantam)	C
Corn (Dixie 17, Funk 134 and 512)	B			Cotton	A
Corn (Spancross)	B			Tobacco	BC
Corn (bantam)	A	Peach	AB	Bean and celery	B
Sweet potato (Triumph)	B	Iris and begonia	B	Cucumber and squash	B
Apricot (Moorpark)	ABC			Squash (Hubbard and zucchini)	AC
Prune (late Italian)	AB			Cabbage, cauliflower, eggplant, onion, pepper, and soybean	A
Peach (some varieties under best conditions)	B	Class 4			
		Azalea	BC	Parsnips (hollow crown)	C
Peach (Elberta, Lovell stock)	C	Begonia (some varieties)	B	Tomato (Marglobe)	C
Strawberry (Marshall)	C	Rose, geranium, coleus, lilac, spirea	B	Chrysanthemum	AB
Grapes (some European varieties)	RW	Verbena	A	Marigold, privet, and snapdragon	A
Iris	AB	Alfalfa	AB	Sweet pea (Spencer hybrid)	C
		Clover	B	Rhododendron (Pink Pearl)	C
		Red clover	C	Rose (Talisman)	C
		Bean (Tendergreen bush)	C	Zinnia	B
		Carrot (Chantenay)	C	Citrus	A

TABLE 4 *(continued)*

Sensitive		Intermediate		Resistant	
		CULTIVATED PLANTS			
		Carrot	A	Laurel and locust	C
		Grape (Concord)	CR	Pine (old needles)	B
		Lettuce	C	Carrot	C
		Sweet potato (Nigger Killer)	B	Petunia	C
		Parsnip, peas, potato, rhubarb, spinach, sugar beet	A		
		Wheat	A		
		Barberry, bridal wreath, honeysuckle, and mock orange	A		
		Apple (Wealthy and Winesap)	C		
		Arbor vitae	C		
		NATIVE PLANTS			
Class 1		Class 3		Class 5	
Pine (ponderosa, young needles)	BC	Pokeweed, crabgrass	B	Pigweed, dock	B
Larch	C	Smartweed, Johnson grass	B	Carpet weed	B
Hypericum	B	Oxalis	B	Pine (lodgepole)	C
		Chickweed, barnyard grass	B	Pine (ponderosa, old needles)	C
Class 2		Class 4		Class 6	
Crabgrass	B	Pokeweed, pigweed, lamb's quarters, dock, plantain	B	Dandelion, plantain, purslane, galinsoga, bidens, nightshade, and ragweed	B
Cattail	B			Douglas fir, grand fir, hemlock, white pine, Engelmann spruce	C
Johnson grass, oxalis, smartweed	B				
Buckwheat	B				
Pine (ponderosa, 3-4-month-old needles)	C				

Source: Thomas & Hendricks (1965). (By permission from "Air pollution handbook", by P. L. Magill, F. R. Holden & C. Ackley, editors. Copyright 1956. McGraw-Hill Book Company, Inc.)

Concentrations required to cause slight injury in 7-9 days: classes 1 and 2 = 5 p.p.t.m. or less; class 3 = 5-10 p.p.t.m.; classes 4, 5, and 6 = more than 10 p.p.t.m.; higher concentrations required proportionally less time.

A = American Smelting and Refining Company; B = Boyce Thompson Institute; C = State College, Washington; R = B.L. Richards; W = F.W. Went.

in 22 hours, the injured tissue containing 297 p.p.m. fluorine. These results have been confirmed by other investigators.

Extensive fumigation experiments with low concentrations of hydrogen fluoride have been carried out by the Boyce Thompson Institute (Zimmerman, personal communication, 1953; Zimmerman & Hitchcock, 1956), Washington State College (Adams, Applegate & Hendrix, the 1957), the Stanford Research Institute (Benedict & Breen, 1955), and the American Smelting and Refining Company (Thomas & Hendricks, 1956). This last-mentioned work has defined the susceptibility of many species to hydrogen fluoride. Table 4 summarizes these results so far as they are available. The Boyce Thompson data are based on seven- to nine-day fumigation periods. If injury occurred with less than 5 p.p.t.m., the plant was considered sensitive; with 5-10 p.p.t.m., intermediate; and with more than 10 p.p.t.m., resistant. The data from other laboratories were evaluated using these criteria, if possible.

It is seen that the number of plants that may be considered to be very sensitive is small. In the field these are usually the only plants that are

TABLE 5

AVERAGE EXPOSURES AND FOLIAR FLUORIDE CONCENTRATIONS AT THREE HYDROGEN FLUORIDE FUMIGATION LEVELS, ALL OF WHICH CAUSED INCIPIENT FOLIAR INJURY IN DAYLIGHT

Plants	1.5 p.p.t.m.		5 p.p.t.m.		10 p.p.t.m.		Average	
	t.c.[a]	p.p.m.[b]	t.c.[a]	p.p.m.[b]	t.c.[a]	p.p.m.[b]	t.c.[a]	p.p.m.[b]
Gladiolus	97	37	119	46	137	57	118	47
Blueberry	97	53	92	72	118	64	103	63
Larch	107	53	119	147	118	106	115	102
Pine, ponderosa, lodgepole, white	438	61	258	77	243	65	313	67
Rhododendron	438	61	258	77	243	65	313	67
Arbor vitae	318	138	286	104	—	—	301	121
Apricot, cherry, peach, prune	246	69	172	79	194	109	187	86
Apples - 4 varieties	302	179	379	183	346	163	343	175
Lilac	138	123	248	216	—	—	193	175
Elm, mulberry, willow, maple	246	217	31[illegible]	231	300	215	280	223
Corn	—	—	747	178	496	47	622	113
Grape	—	—	252	138	344	122	298	130
Squash	—	—	631	134	421	179	526	157
Sweet pea	313	327	307	148	374	141	335	205
Raspberry	306	243	306	216	183	162	265	207
Pepper	218	149	274	203	188	244	227	199
Tomato	—	—	327	278	302	207	315	243
Carrot	284	323	398	723	425	307	369	451

Source: Adams, Applegate & Hendrix (1957).

[a] Hours of exposure time X p.p.t.m. hydrogen fluoride.

[b] Fluoride in leaves, dry basis.

injured. There may be a large range of susceptibility among varieties of a single species. This is true of gladiolus, grape, apricot, corn, and sweet potato. In some of these plants the most sensitive variety may absorb less fluoride than the resistant variety. For example, the Algonquin variety of gladiolus with 7 % leaf injury absorbed 611 p.p.m. fluorine, while the Shirley Temple variety with 54 % injury absorbed only 138 p.p.m.

Table 5 is a rearrangement of fumigation data by Adams et al. (1957) giving (*a*) the " exposure factor " as the summation of the number of hours of daylight fumigation times the concentration of hydrogen fluoride in parts per thousand million, and (*b*) the fluoride content of the foliage in parts per million dry basis, for 29 different plants at the time when incipient injury occurred. Other plants that were not injured are not included in this table.

TABLE 6

AVERAGE EXPOSURES AND FOLIAR FLUORIDE CONCENTRATIONS AT TWO HYDROGEN FLUORIDE FUMIGATION LEVELS WHICH CAUSED INCIPIENT FOLIAR INJURY IN THE DARK

Plant	1.5 p.p.t.m.		5 p.p.t.m.		Average	
	t.c. *a*	p.p.m. *b*	t.c. *a*	p.p.m. *b*	t.c. *a*	p.p.m. *b*
Gladiolus	82	59	122	44	102	52
Blueberry	82	34	288	103	185	69
Larch	82	62	83	73	83	68
Pine, ponderosa, lodgepole, white	—	—	222	61	222	61
Apricot, cherry, peach, prune	320	69	294	74	307	72
Apple - 3 varieties	336	72	486	106	406	89
Elm, mulberry, willow	115	68	292	85	204	77
Grape	362	51	522	84	442	68
Raspberry	163	88	—	—	163	88
Pepper	188	136	—	—	188	136
Carrot	188	250	430	309	309	280

Source: Adams, Applegate & Hendrix (1957).

a Hours of exposure time X p.p.t.m. hydrogen fluoride.
b Fluoride in leaves, dry basis.

Table 6 contains similar data for fumigations in the dark. The 1.5 p.p.t.m. fumigations were applied eight hours per day, five days each week. The 5 p.p.t.m. and 10 p.p.t.m. fumigations were applied twice a week, eight and four hours per day respectively. The exposures (Σ c.t.) were thus approximately equal in the three levels of fumigation. All fumigations were done in the day-time, but the cabinets were masked for the dark fumigations.

The most sensitive plants, such as gladiolus, had small exposure factors, and small amounts of fluoride in the leaves caused injury. The opposite was true of the most resistant plants. Some species required a large exposure factor to take up the small amount of gas that caused injury, indicating a very slow rate of uptake, but considerable sensitivity to the absorbed fluoride. Corn is an extreme example of this type of plant. It exhibits a special type of lesion as already described. The different varieties of corn show a wide range of sensitivity (see Table 4), indicating possible different rates of uptake of the gas. The pines behave similarly. Young pine needles, which are very sensitive to hydrogen fluoride, probably take up the gas more rapidly than older needles.

Adams et al. (1957) made the following additional observations which are statistically significant:

(1) Injury appeared on the various species at about half the foliar fluoride level in the dark as in the light, but the exposure factor necessary to cause injury in the dark was 91 % of that in the light.

(2) Injury appeared in the 1.5 p.p.t.m. fumigation with about 70 % of the exposure factor that caused injury in the 5 p.p.t.m. fumigation. The latter, in turn, required 107 % of the 10 p.p.t.m. exposure factor to cause injury.

Photosynthesis and respiration

In view of the great susceptibility of some species and the resistance of others, it is important to know whether fluoride interferes with the physiological activity of the plant when it does not cause visible lesions. An excellent yardstick for such activity is furnished by measurement of photosynthesis and respiration of the plants before, during, and after the fumigation treatment. This is readily accomplished with the automatic carbon dioxide analysers. Preliminary work on these measurements was carried out by the author and R. H. Hendricks at the American Smelting and Refining Company (Thomas, 1956; Thomas & Hendricks, 1956).

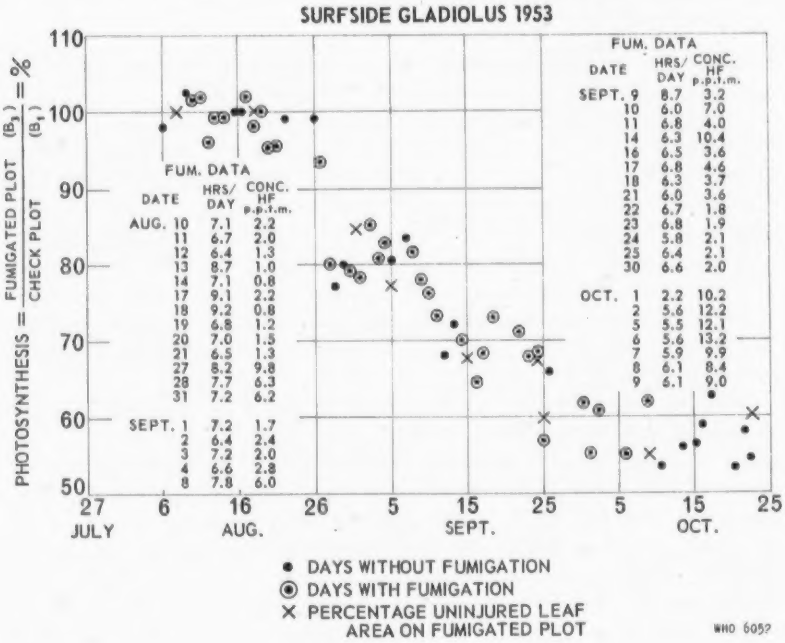
Fumigation of several plant species with hydrogen fluoride with simultaneous measurement of the carbon dioxide exchange indicated that there was a threshold concentration for each plant below which no interference with photosynthesis occurred, and above which photosynthesis was affected, even though no lesions were produced on the leaves. In an experiment with Surfside gladiolus, fumigations were applied at 1-10 p.p.t.m., about seven hours per day and five days per week for nearly two months. Injury to the blades was produced starting at the tip and extending gradually down the blade until 40 %-45 % of the area was destroyed. The assimilation rate fell off at the same rate as the leaf area was destroyed (Fig. 1) until it was reduced by 38 %-46 %. Evidently the green tissue remained fully functional until it was killed as shown by the advancing necrotic front, suggesting the absence of invisible injury even though a great deal of visible injury occurred.

Consideration of the fumigation details listed in Fig. 1 indicates that there was a sudden 12 % decrease in the rate of photosynthesis on 28 August following a concentration of 9.8 p.p.t.m. on 27 August. With the fumigations remaining between 2 and 7 p.p.t.m. during the subsequent two weeks, this excessive decline was arrested and a slope corresponding to the rate of increase in leaf destruction was re-established. A smaller similar effect followed the 10.4 p.p.t.m. fumigation on 14 September. The leaf area destroyed did not increase appreciably after 25 September, and even the 10-13 p.p.t.m. fumigations after 1 October had little effect on either photosynthesis or leaf destruction. Possibly the resistance of the plant was increased either by maturity or low temperatures at that time. Similar results were obtained in several other comparable experiments with gladiolus. It is likely that the threshold for this variety of gladiolus is about 6-7 p.p.t.m. in August and September, certainly less than 10 p.p.t.m., with these intermittent fumigations.

A number of fumigations were carried out with several crops, using subthreshold concentrations of hydrogen fluoride. These are summarized in

FIG. 1

DAILY CARBON ASSIMILATION BY A GLADIOLUS PLOT
FUMIGATED WITH HYDROGEN FLUORIDE
EXPRESSED AS PERCENTAGE OF A CONTROL PLOT,
COMPARED WITH REDUCTION OF LEAF AREA DUE TO FUMIGATION



FUM. DATA

DATE	HRS/ DAY	CONC. HF p.p.t.m.
AUG. 10	7.1	2.2
11	6.7	2.0
12	6.4	1.3
13	8.7	1.0
14	7.1	0.8
17	9.1	2.2
18	9.2	0.8
19	6.8	1.2
20	7.0	1.5
21	6.5	1.3
27	8.2	9.8
28	7.7	6.3
31	7.2	6.2
SEPT. 1	7.2	1.7
2	6.4	2.4
3	7.2	2.0
4	6.6	2.8
8	7.8	6.0

FUM. DATA

DATE	HRS/ DAY	CONC. HF p.p.t.m.
SEPT. 9	8.7	3.2
10	6.0	7.0
11	6.8	4.0
14	6.3	10.4
16	6.5	3.6
17	6.8	4.6
18	6.3	3.7
21	6.0	3.6
22	6.7	1.8
23	6.8	1.9
24	5.8	2.1
25	6.4	2.1
30	6.6	2.0
OCT. 1	2.2	10.2
2	5.6	12.2
5	5.5	12.1
6	5.6	13.2
7	5.9	9.9
8	6.1	8.4
9	6.1	9.0

TABLE 7

EFFECT OF FUMIGATION WITH RELATIVELY LOW CONCENTRATIONS OF HYDROGEN FLUORIDE ON THE PHOTOSYNTHESIS OF PLANTS

Plot	Plant	HF fumigation			Photosynthesis $\left(\frac{\text{Fum. Plot}}{\text{Check Plot}}\right)$ Reduction due to fumigation %	Leaf area destroyed %
		Duration[a] (hours)	Concentration			
			Maximum p.p.t.m.	Average p.p.t.m.		
1950-B3	Fruit trees	183	9.0	2.6	14	10
1950-B4	" "	39	15.0	4.8	2	< 5
1951-B3	" "	8	4.0	3.9	0	0
1952-C3	Gladiolus, Surfside	53	11.3	6.5	15	15
1952-C4	" "	44	9.8	6.4	28	30
1953-B3	" "	205	13.2	4.5	45	44
1953-D3	" "	128	33.4	5.7	20	22
1953-A1	" Alladin	30	5.1	3.9	0	3
1953-C2	" Algonquin	111	7.1	4.3	2	3
1953-D4	" Commander " Koehl	42	10.2	5.5	0	3
10-2	Mixed grain	10	200	145	0	Trace
1951-D4	Cotton	77	692	237	0	Trace
1952-A4	Cotton	138	33	17	0	0

[a] Fumigations 4-9 hours per day.

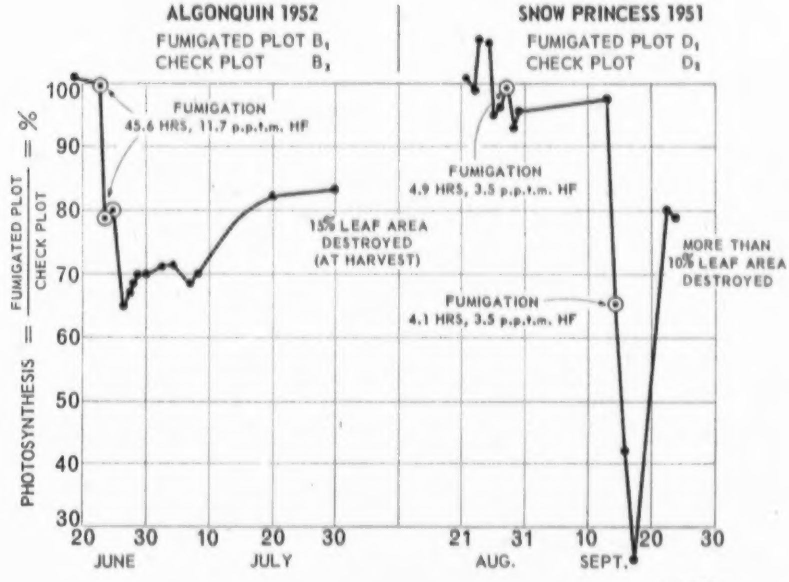
Table 7. It is evident that reduction in photosynthesis and leaf destruction are nearly equal so long as the threshold is not exceeded.

When higher concentrations of hydrogen fluoride were used on gladiolus, effects like those shown in Fig. 2 were obtained—viz., a sharp reduction in the photosynthetic level followed by a rise to the level anticipated from the amount of acute injury incurred. Similar curves have been obtained with alfalfa fumigated with 250-1000 p.p.t.m. for a few hours (Fig. 3), also with young barley and fruit trees at about 50 p.p.t.m. With cotton, concentrations up to 1100 p.p.t.m. had only a very slight effect on photosynthesis, in accord with the great resistance of this plant (Fig. 4). As much as 5000 p.p.m. fluoride has been found in the leaves of apparently uninjured cotton.

The reduction of photosynthesis below the level expected from the amount of leaf destruction can properly be called " invisible injury ". The effect is similar to that shown in comparable sulfur dioxide fumigations except that the recovery from the high fluoride treatment is slower than from the sulfur dioxide. In the latter case it is only necessary to oxidize the absorbed sulfite to sulfate to remove the toxic material, a process that usually reaches completion in the leaves in a few hours. Fluoride toxicity in the tissues must be reduced by translocation of the element to limited areas of concentration in the margins and tips of the leaves which may be killed, while

FIG. 2

EFFECT OF RELATIVELY LOW CONCENTRATIONS OF HYDROGEN FLUORIDE ON THE TOTAL DAILY CARBON ASSIMILATION OF GLADIOLUS



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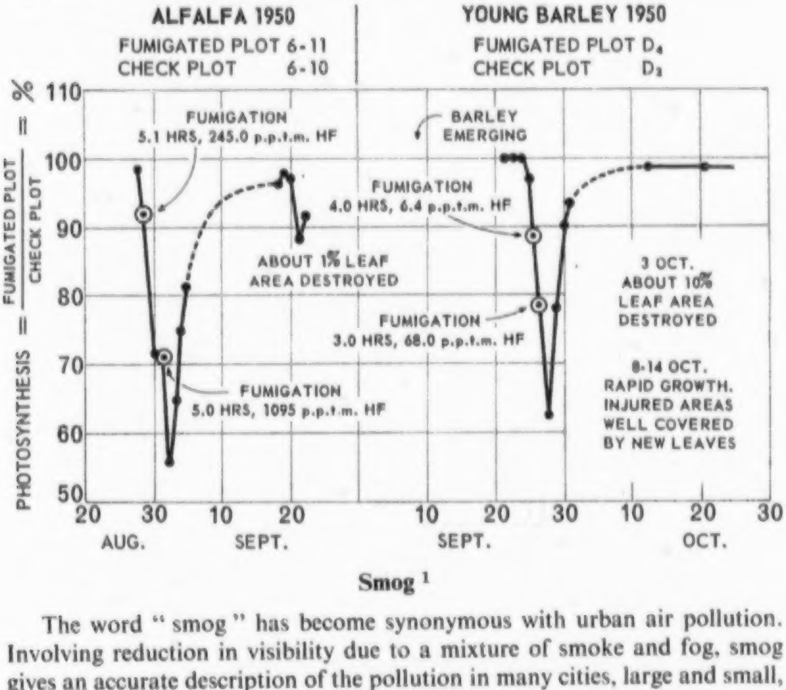
only non-toxic concentrations remain in the body of the leaves. This requires time. Table 8 gives an analysis of the data for the fumigations with high concentrations that caused interference with the photosynthesis of the unmarked areas of the leaves. Complete recovery from these temporary effects of the gas required from 4 to 25 days and was equivalent to total cessation of assimilation for 0.2 to 3.8 days. The latter estimates were made by finding the average suppression of assimilation below that indicated by the leaf destruction and multiplying by the time required for complete recovery. They actually represent only a few per cent of the total photosynthesis of the crop. Multiple severe fumigations might cause extensive interference with the plant growth.

It is evident that a satisfactory basis for estimation of crop losses due to fluorides might be established on the principles suggested in the foregoing work. With knowledge of the fluoride concentrations in the field and the threshold values for the crops concerned, visible and invisible injury could be evaluated. This would entail intensive experimental work with the various crops. Fortunately only a few crops are sensitive in the parts per thousand million range, so that the problem would be definitely limited.

FIG. 3

EFFECT OF RELATIVELY HIGH CONCENTRATIONS OF HYDROGEN FLUORIDE ON THE TOTAL DAILY CARBON ASSIMILATION OF ALFALFA AND BARLEY



Smog[1]

The word " smog " has become synonymous with urban air pollution. Involving reduction in visibility due to a mixture of smoke and fog, smog gives an accurate description of the pollution in many cities, large and small, which burn coal and have a good deal of fog, e.g., London. Appreciable levels of sulfur dioxide and other gaseous products due to incomplete combustion of the coal are generally present together with soot, grime, and ash. In the lethal London smog of December 1952 (Wilkins, 1954), maximum values of 1.34 p.p.m. sulfur dioxide and 4.5 mg/m^3 of aerosols, as averages of the peak 48-hour sampling period, were measured. There is a wealth of data on the levels of these two constituents as they have appeared in many English and American cities, but there are hardly any determinations of other pollutants such as sulfuric acid aerosol, oxides of nitrogen, fluorides, and organic compounds—all of which might be very important.

The London smog

The phytotoxicity of London smog is generally believed to be due largely to its sulfur dioxide content. Metcalfe (1953) stated that although the ever-

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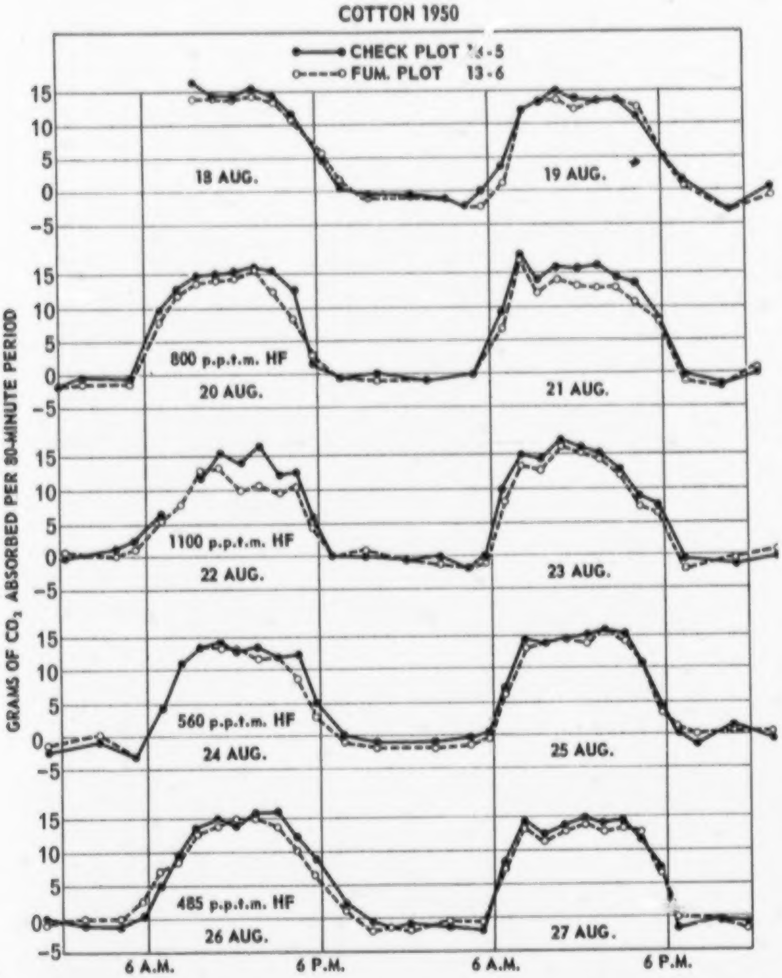
[1] For illustrations of typical lesions on a number of plants caused by ethylene, photochemical smog (probably peroxyacetyl nitrite, PAN) and ozone, see Plates 3 and 4 (facing pages 264 and 272).

FIG. 4

PHOTOSYNTHESIS AND RESPIRATION OF COTTON OVER A 10-DAY PERIOD DURING WHICH FOUR FUMIGATIONS WITH HYDROGEN FLUORIDE WERE APPLIED



greens near Kew Gardens had become stunted owing to deposition of soot on the leaves, as some older work by Rushton (1921), Rhine (1924), and many others would suggest, the more acute symptoms of ill health were due to toxic substances in the atmosphere rather than to reduction of the light by the surface deposit.

TABLE 8

EFFECT OF FUMIGATION WITH RELATIVELY HIGH CONCENTRATIONS OF HYDROGEN FLUORIDE ON THE PHOTOSYNTHESIS OF PLANTS

Plot	Plant	HF fumigation			Decrease in photosynthesis $\left(\frac{\text{Fum. Plot}}{\text{Check Plot}}\right)$ Due to fumigation				Leaf area destroyed %
		Duration [a] (hours)	Conc.		Temporary			Permanent	
			Maximum p.p.t.m.	Average p.p.t.m.	Duration (days)	Average [b] %	No. days lost	%	
A1	Fruit trees	4	118	52	18	11	2.0	28[c]	50[d]
A3	" "	13	-22	18	25	15	3.8	35	22[e]
D1	Gladiolus, Snow Princess	4	40	29	9	29	2.6	18	> 10
B1	" Algonquin	46		12	21	12	2.5	17	15
C3	" Surfside	53	11	7	25	8	2.0	15	15
D4	Young barley	7	68	37	7	15	1.1	1[f]	10
6-11	Alfalfa	10	1 020	630	14	15	2.1	2	1
13-6	Cotton	20	1 100	740	4	6	0.2	0	0

[a] All fumigations except B1 applied 4-7 hours per day.
[b] Temporary reduction below level of permanent injury.
[c] Some new growth of leaves reduced percentage loss.
[d] Approximate value 10 days after fumigation before new growth.
[e] Estimated before leaf fall.
[f] Rapid new growth restored activity.

> Examples of these acute symptoms are the shedding of flowers and leaves of begonia and many other plants... the blackening and death of buds and flowers of certain orchids, notably Calanthes, the scorching of the foliage of cinerarias and primulas and the almost complete destruction of such plants as *Coleus blumei* ...

Experimental fumigations at Kew with low concentrations of sulfur dioxide comparable to concentrations in London air are said to give symptoms similar to those exhibited by the same species during foggy weather " provided the atmosphere is sufficiently moist and the temperature sufficiently high". The severity of the effects increased with increasing sulfur dioxide concentration.

The question arises whether the ignoring of other constituents of the smog mixture is justified. It should be pointed out that some of the symptoms described are more characteristic of other pollutants such as ethylene, organic bases such as acridine or perhaps hydrogen fluoride than of sulfur dioxide. However, as Metcalfe suggested, the latter doubtless is a very important contributor to the phytotoxicity of the London smog because of its high concentration.

Bleasdale (1952a, b) in Manchester worked with a rye grass *Lolium perenne* (Aberystwyth S 23) and observed a definitely retarded growth in the open atmosphere or in the greenhouse as compared with a greenhouse supplied with air scrubbed by water sprays. No evidence of acute leaf damage was seen in the greenhouse with unscrubbed air, but the rate of senescence of the plants was greater than in washed air. Rate of tiller formation and number of leaves as well as dry weight were reduced by the pollution. Evidently the processes of cell division and cell development are affected along with photosynthesis. In one experiment the pollution reduced the numbers of tillers and leaves by 54% and 44% respectively, while the dry weight was reduced by 33%.

The fertility of the soil was also important. In one of the local infertile soils, polluted air reduced by 37% the dry weight of plants grown in April and May. After addition of mineral fertilizer to this soil, the same yield was obtained in polluted air as in scrubbed air. When the experiment was repeated in June and July, the corresponding dry weights in the polluted air were reduced by 57% and 33%, respectively, below those in scrubbed air. The average daily sulfur dioxide concentrations were approximately equivalent in all the foregoing experiments, and nearly all were less than 0.1 p.p.m. In the last experiment, the relative humidity of the polluted air was raised by a spray and some sulfur dioxide was washed out.

In a final experiment with rye grass, carried out from November 1951 to February 1952, some of the plants were in either scrubbed or polluted air only during the night (4.30 p.m.-8.30 a.m.). The location of these plants was reversed during the day. Other plants were in either scrubbed or polluted air continuously. The dry weights of the four groups are given in Table 9.

Table 9 indicates that it is advantageous to expose the plants to unscrubbed polluted air during the day or during the night, but not continuously. The amount of dead tissue on the plants at harvest was only 6%-8% of the total dry matter. The effect of the pollution in hastening senescence was therefore slight, though large with regard to percentage.

The sulfur dioxide concentrations in this experiment were considerably higher than in the others. The average for 118 days covering this period was 0.12 p.p.m. with six days between 0.3 and 0.5 p.p.m. In the unscrubbed greenhouse, with its humidifying sprays, the average for 114 days was 0.06 p.p.m. with five days between 0.2 and 0.3 p.p.m. The reason for the increased growth of the plants exposed intermittently to unscrubbed air is not apparent. Increased growth in sublethal sulfur dioxide fumigations has not been observed except when the plants were sulfur deficient.

TABLE 9

YIELD OF RYE GRASS AFTER VARIOUS DAY (D) AND NIGHT (N) EXPOSURES TO SCRUBBED (S) AND POLLUTED (P) AIR DURING WINTER OF 1951-52

Day treatment	Night treatment			
	SN	PN	SN	PN
	Green tissue		Dead tissue	
	Dry weight mg/plant			
SD	53 ± 3	62 ± 3	3.3 ± 0.2	4.7 ± 0.2
PD	62 ± 3	44 ± 3	4.1 ± —	4.1 ± 0.1
	Change due to pollution, %			
$\frac{\text{SD, PN — SD, SN}}{\text{SD, SN}}$	+ 17		+ 43	
$\frac{\text{PD, SN — SD, SN}}{\text{SD, SN}}$	+ 17		+ 24	
$\frac{\text{PD, PN — SD, SN}}{\text{SD, SN}}$	— 17		+ 24	

Source: Bleasdale (1952 a, b).

Mention has already been made of Bleasdale's explanation for the apparently beneficial effect of intermittent exposures—viz., the sulfurous acid stimulates the production of sulfhydryl groups but suppresses their activity if present continuously. When the sulfur dioxide is removed, the elevated level of sulfhydryl groups results in increased cell division and growth.

The possibility remains that other pollutants in addition to sulfur dioxide participated in producing the effects described. It would be of interest to repeat the experiments, supplementing them by the use of carbon-filtered air, with and without known concentrations of sulfur dioxide.

PLATE 3

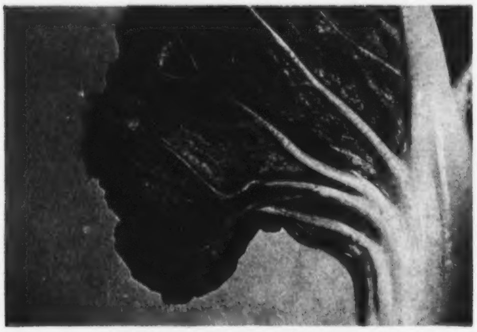


Fig. 9. Silver leaf on lower surface of Swiss chard due to photochemical smog

Fig. 10. Silver leaf on lower surface of spinach due to photochemical smog

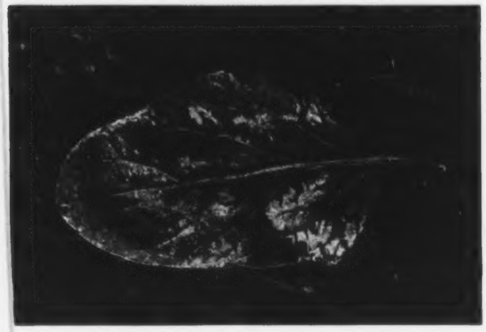


Fig. 11. Severe lower surface smog damage to spinach

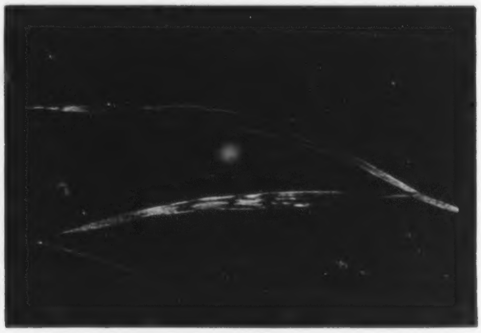


Fig. 12. Bands on *Avena fatua* (wild oats) from multiple exposures to photochemical smog

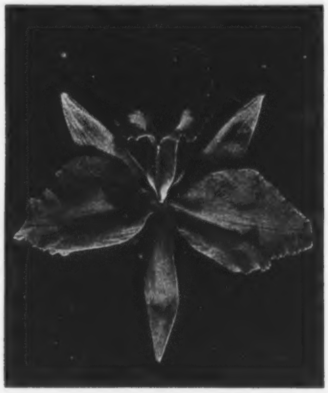


Fig. 13. Smog damage on orchid (dry sepal) probably due to ethylene

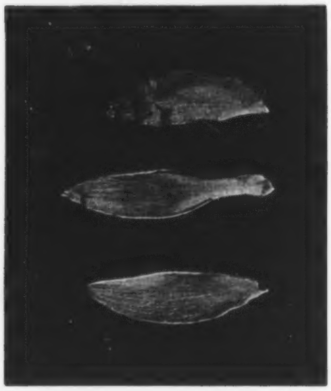


Fig. 14. Dry sepals of orchids probably due to ethylene

Acknowledgement is made on page 275 to the persons who furnished the above coloured photographs.

Los Angeles type smog

The Los Angeles type smog is radically different from the London type. In fact, the word " smog " is a misnomer in the case of Los Angeles because, usually, neither ordinary smoke nor fog is present, owing to the use of smokeless fuels in an essentially desert area. The pollution of this area arises largely from photochemical reactions between oxides of nitrogen and organic vapours, both derived from the incomplete combustion processes of industry, automotive traffic and incinerators, and also from gasoline evaporated directly into the atmosphere (Haagen-Smit, 1952; Haagen-Smit & Fox, 1956). Of course, many other inorganic and organic substances are present in addition (Los Angeles Air Pollution Control District, 1951; Stanford Research Institute, 1954).

The phytotoxicity of this smog is very striking, particularly on the so-called " salad crops "—i.e., spinach, romaine, endive—and also on table beets, Swiss chard, and celery. Characteristic lesions are produced. First there is a silvering or bronzing of the lower epidermis, due to collapse of the sub-epidermal cells. Later the injury may extend through the leaf, giving white or spotted collapsed areas on the upper surface. An excellent group of photographs in colour, illustrating smog injury, is given by Middleton et al. (1956).

The phytotoxicants in the smog have been under investigation for many years. Apart from ozone, which, as is indicated later, does not cause typical smog lesions, none were found which produced lesions at the concentrations present in the smog. It was assumed that the smog effects were caused by the mixture of oxidants. Even Compound X, peroxyacetyl nitrite (or nitrate), or PAN, prepared by the Franklin Institute (Stephens et al., 1956a, b), did not appear to be highly phytotoxic. This substance, identical in infra-red spectrum with that isolated from smog (Stephens et al., 1960), has been prepared in quantity by photochemical reaction between n-butene and nitrogen dioxide, purified by freeze out and gas chromatographic techniques, then diluted with nitrogen to known concentration and stored under pressure for controlled fumigation experiments. It has a half-life of about 13 hours. It is a powerful eye irritant. Plant studies with PAN (Taylor et al., 1960) indicate great phytotoxicity for herbaceous plants such as pinto bean, the " salad crops ", petunia and many others. Typical smog-type lesions on petunia are produced by 0.05 p.p.m. in about four hours. Evidently this compound is a significant component of the smog mixture.

Bobrov has studied the injury process in detail, using hand-sectioning of the fresh material and staining by thionine and other dyes to facilitate the observations (Bobrov, 1952a, b, 1955; Glater, 1956; Los Angeles Country Air Pollution Control District, 1951). Her description is of great interest. As the first response,

> ... the lower epidermis is raised in tiny blisters, which, on close inspection, may be seen with the naked eye. In the region of some, but not all of the stomata, the guard cells together with several surrounding epidermal cells are engorged with water and push up above the sub-stomatal chamber, thus forming a blister ... The epidermal cells swell in length and width, become turgid and the walls lose their fluted appearance and become regular. The guard cells increase in width but not in length, opening the stomata wide. The attached epidermal cells then collapse.

The cells lining the lower and upper sub-stomatal chambers are the first to plasmolyse. Then the stomata probably close.

Annual bluegrass *(Poa annua)* is one of the plants most sensitive to smog, and its general distribution has made it very useful as an indicator of smog. Bobrov (1955) presents a detailed anatomical study of the plant showing its open structure with stomata on both sides of the leaves and indicating the sensitive areas where tan spotting or transverse banding occurs. These are limited to those cells that have just completed maximum expansion. They are located at the tip of the youngest leaf and progressively farther down the blades of the older leaves. Initial and most serious damage occurs around the sub-stomatal chambers. Chloroplasts disintegrate, plasmolysis follows, and, ultimately, total dehydration of damaged cells results in " mummification " of the mesophyll tissue in the affected areas.

Noble (1955) has generalized the observations concerning the banding on *Poa annua*. In all but one of 50 species studied, even on broad-leaved plants, some form of banding occurred following a single exposure to smog. Repetition of the fumigation on successive days resulted in a series of bands progressively farther down the leaf. As many as four bands on a single leaf of petunia or mimulus were easily produced. The injured area was always that in which cellular differentiation had most recently occurred, as shown by Bobrov (1952a, b), who also suggested that the increased resistance to the smog shown by the older leaf surfaces was due to suberization of those surfaces after differentiation was completed, as described by Scott (1950).

Table 10 is a list of plants prepared at the Citrus Experiment Station in Riverside, California, classifying many species and varieties in the Los Angeles area as sensitive or resistant to smog. The classification in this table is well confirmed by other observers (Thomas & Hendricks, 1956).

A specific test for smog injury was suggested by Nielson, Benedict & Holloman (1954), who observed that the smog-injured areas start to fluoresce bright bluish-white under a mercury lamp several hours after fumigation and continue to fluoresce for about a week. The test is not given for gas injury due to sulfur dioxide, hydrogen fluoride, or chlorine, etc.

Smog injury exhibits several degrees of severity, which are called transient, chronic, and acute. The first is a temporary water-soaked condition, which soon clears up without producing permanent lesions. The second

TABLE 10
SENSITIVITY OF PLANTS TO INJURY BY SMOG IN SOUTHERN CALIFORNIA

Susceptible	Resistant
TREE CROPS	
Apricot, Grapefruit, Walnut	Almond, Apple, Cherry, Date, Grapefruit, Lemon, Olive, Orange, Peach, Plum
FIELD CROPS	
Alfalfa, Oats, Sudan grass, Sugar beet	Sweet clover, Barley, Mustard (black and white), Wheat, Blackeyed bean, Vetch
VEGETABLE CROPS	
Bean, common (Golden cluster, Pink, Pinto, Small white), Bean, lima (Fordhook 242), Beet, Endive, Lettuce (Romaine), Spinach, Swiss chard (Lucullus), Celery, Onion, Parsley, Parsnip, Turnip, Grape (Carignane, Mataro, Mission, Palomino, Pedro Ximenes, Zinfandel)	Bean, common (Bountiful, Kentucky Wonder), Bean, lima (Concentrated Fordhook, Westan), Chinese cabbage, Corn, Eggplant, Lettuce (head), Mustard, Pea, Radish, Rhubarb, Tomato, Leek, Pepper, Potato, Squash, Swiss chard (large ribbed), Broccoli, Cabbage, Cauliflower, Muskmelon, Grape (Concord)
ORNAMENTAL CROPS	
Petunia, Chrysanthemum (some varieties), Grass, annual rye (Perennial rye), Snapdragon, Larkspur, Ageratum, Carnation, Eugenia, Orchid, Palm Kentia), Pansy, Pepper, Rose, Zinnia	Acacia, Calendula, Lobelia, China aster, Chrysanthemum (most varieties), Sweet pea, Grass, Bermuda (Kentucky blue), Pansy, Stock, Eucalyptus, Dahlia, Gaillardia Hypericum Geum, Viola, Ivy, White clover, Oak, Palm (Plume, Washington)
SMALL FRUIT	
	Blackberry, Boysenberry, Currant, Loganberry, Strawberry, Youngberry

Source: Kendrick et al. (1956).

is a mild degree of injury with effects resembling chronic sulfur dioxide markings. In addition, there appears to be invisible injury. When acute injury occurs on the " salad crops " they are rendered virtually worthless commercially. Middleton, Kendrick & Schwalm (1950b) estimated that the loss for 1949 in Los Angeles County alone was US $500 000. Middleton et al. (1956) estimated the annual loss in 1953 and 1954 in the Los Angeles area at over US $3 000 000 and, in 1956, in excess of US $5 000 000 (Middleton et al., 1957). The 1956 loss in the San Francisco Bay area was US $1 000 000 (Middleton, Darley & Brewer, 1957). No estimate was made of suspected " invisible " injury due to smog to such plants as tomato, sugar beet, and avocado.

Effects of ozone

Ozone is an important constituent of Los Angeles smog, usually comprising more than half the total oxidants. It is also phytotoxic in exposures of a few hours at about 0.2 p.p.m. Injury due to ozone (Haagen-Smit et al., 1952; Middleton et al., 1955; Richards, Middleton & Hewett, 1958), is quite different from typical smog injury. The lesions are generally confined to the upper surface. They may be uniformly distributed white or brown flecks or stipples, or irregularly distributed blotches. Histologically they represent a bleaching or browning of the upper ends of some palisade cells without involving the upper epidermis.

Ozone injury appears to be much more widespread than the earlier observations suggested (Taylor et al., 1960). Close examination of leaves having typical smog damage usually reveals some ozone-type injury also. Ozone flecking is widely distributed on grape, citrus, avocado and other broad-leaf woody plants in the outlying areas of the Los Angeles Valley and surrounding hills. Ponderosa pine needles appear to be similarly affected. Extensive ozone flecking on tobacco has recently been observed in the eastern Atlantic states of the USA and in Canada (Heggestad & Middleton, 1959). Flecking on white pine is also observed even in places far removed from urban and industrial sources of pollution, possibly as a result of photochemical reactions involving volatile organic substances derived from vegetation (Went, 1955).

Effects of acid aerosols

A " pock mark " type of injury (Middleton et al., 1957; Thomas et al., 1952), particularly on the upper surfaces of table beets and Swiss chard, has been ascribed to acid aerosols associated with fog. Evidently the fog droplets which settle on the leaves contain enough acid, and perhaps other toxicants also, to injure the leaf. When indicator paper was pressed against such leaves, a pH of 3 or less was often observed. Injury appeared after the moisture evaporated. This type of injury needs more critical study.

Effects of ethylene

Injury to orchids by ethylene in the smog has been reported to have resulted in large monetary losses in Los Angeles. The gas causes the sepals in many Cattleya and Phalaenopis flowers to wither and dry at a concentration of 5 p.p.t.m. (Middleton et al., 1956), while other parts of these plants can withstand much higher concentrations without any injury.

Ethylene causes epinasty and inhibition of nutation in marigold, sweet pea, and tomato leaves at 0.05-0.10 p.p.m. (Crocker, Hitchcock & Zimmerman, 1935; Crocker, Zimmerman & Hitchcock, 1932; Middleton et al., 1956). The gas interferes with the opening of carnation flowers and causes dropping of snapdragon flowers. At 0.4 p.p.m. it produces leaf irregularities and poor flower formation in tulips and narcissus.

Ethylene is difficult to measure in the air, and little is known of its concentration in polluted atmospheres. It has given concern principally in greenhouses that had leaking gas pipes (Hitchcock, Crocker & Zimmerman, 1932, 1934; Zimmerman, Hitchcock & Crocker, 1931). Economic injury under these conditions has been reported in lilacs, narcissus, tulips, and roses.

Nitrogen oxides

The concentration level of nitrogen oxides in the atmosphere is probably always too low to cause plant damage. Benedict & Breen (1955) found that their most sensitive weeds required at least 20 p.p.m. and the most resistant, 50 p.p.m. Middleton et al. (1957) reported that symptoms on pinto beans resembling those caused by sulfur dioxide were produced by 3 p.p.m. nitrogen dioxide in four to eight hours, while complete damage occurred with 30 p.p.m. in two hours.

Fumigation experiments

Haagen-Smit et al. (1952) noted reduced growth of plants due to smog exposures that caused no visible lesions. Hull & Went (1952) made similar observations on alfalfa, endive, oats, spinach, sugar beet and tomato. They also suggested a sensitive test for smog based on the fact that the bending of the avena coleoptile in the regular growth hormone assay is inhibited by smog. Koritz & Went (1953) noted stomatal closure with reduced water uptake and transpiration in tomato plants two to three weeks old as an immediate response to six daily fumigations for one hour, each with 1-n-hexene and ozone at 0.1 p.p.m. oxidant. Reduction of height, leaf area, fresh weight and dry weight occurred, generally with no visible lesions. Decrease in dry weight was as much as 25 %. Evidently, this is " invisible " injury. Cann, Noble & Larson (1954) found characteristic smog effects on plants treated with automobile exhausts to which ozone was added.

The use of a mixture of ozone and olefin, such as 1-n-pentene, 1-n-hexene, or even gasoline, in controlled fumigation studies simulating smog is justified by many observations, indicating that the lesions produced are indistinguishable from those caused by natural smog at the same oxidant level.

Middleton and co-workers at Riverside, California, have made extensive use of the pinto bean as a test plant for smog (Erickson & Wedding, 1956;

Kendrick et al., 1956; Kendrick, Middleton & Darley, 1954; Middleton, 1956; Middleton et al., 1956; Middleton et al., 1957; Middleton et al., 1955; Middleton, Kendrick & Schwalm, 1950a, b; Taylor et al., 1957; Todd, 1955, 1956; Todd, Middleton & Brewer, 1956; Wedding & Erickson, 1955). Two weeks after emergence from the soil, the two primary leaves of the plant are uniformly and highly susceptible to smog damage and remain as sensitive for three weeks longer. Damage is estimated visually on a scale of 10—from no effect at 0 to complete destruction at 10.

These studies have shown the following:

(1) At 0.2 p.p.m. oxidant concentration, as measured by potassium iodide, ozonated hexene causes about the same amount of damage to pinto beans as ozone alone, but at 0.4 p.p.m. ozone is much more reactive (Middleton, 1956).

(2) Ozone damage has only rarely been found in the field (Middleton et al., 1955). The addition of 0.25 p.p.m. ozone to carbon-filtered native air or to unfiltered air of low natural oxidant level causes typical ozone injury to pinto beans. However, addition of this amount of ozone to native air of elevated oxidant level increases the smog damage and an appreciable amount of typical ozone-type injury does not occur. Presumably the ozone content of the air on days of low oxidant level may not be high enough to be toxic, whereas with high oxidant levels the ozone reacts preferentially with other compounds in the air to form additional toxic smog compounds.

(3) Photosynthesis and respiration of *Lemna minor* (duckweed) was studied in the Warburg apparatus by Erickson & Wedding (1956). Ozonated hexene was prepared by mixing 1 volume dilute ozone equivalent to 1 p.p.m. in final dilution with 9 volumes of dilute hexene vapour. The oxidant concentration of the mixture was 0.2 p.p.m. The gas was passed through the Warburg flasks at 250 ml per minute for periods up to 24 hours. In other experiments the same amount of ozone was used without hexene. The plants were severely injured during exposure, with loss of chlorophyll. Fronds separated and droplets of liquid appeared on their surfaces. Eventually there was collapse and discoloration. However, normal growth of the frond primordia resumed in a few days in the absence of smog.

In 24 hours, the ozone-hexene mixture reduced photosynthesis and the chlorophyll content in the plants to 30 % and 50 % of the control, respectively. Ozone alone reduced them both to 60 % of the control. Hexene alone had no effect. Respiration was increased by ozone-hexene to about 115 % during the first 12 hours, then it fell gradually to 90 % during the second 12 hours.

Ozone alone caused only a slight increase of respiration during the first four hours, then a gradual decrease to 90 %. The semi-permeability retention was measured by reading the conductivity of the cell suspension. It was reduced to about 70% of the controls by both gas mixtures. Evidently the lesions and loss of chlorophyll can explain most of the effects noted—except the excess reduction in photosynthesis over chlorophyll, which might be considered invisible injury.

(4) In other studies (Wedding & Erickson, 1955), the retention of radiophosphate by bean plants was measured to ascertain the effect of fumigation treatment with ozonated hexene. During the first one to two hours of the fumigation, sand-cultured plants containing ^{32}P in the leaves, showed a decrease of permeability of the cells, so that disks cut from the leaves lost less phosphate than the controls. Later permeability was increased. Visible damage to the leaves was associated with this increase in permeability.

(5) Ozonated hexene and ozone both stimulated respiration of lemon fruits (Todd, 1956), the former by as much as three-and-one-half times the control, the latter by less than double. The dark green fruits were stimulated the most, ripe yellow fruits the least.

(6) Avocado seedlings were fumigated with ozonated hexene at 0.17 p.p.m. oxidant level for seven hours per day, five days per week for eight weeks, or a total of 280 hours (Taylor et al., 1957). When the plants were not being fumigated they were placed along with the controls in a greenhouse provided with carbon-filtered air. Smog lesions started to develop in two weeks as small bronze or brown spots on the lower surface of fully expanded leaves. This injury increased but did not extend to the upper surface. Some leaves on one-third of the plants developed tip and marginal necrotic areas resembling drought or salt injury. A general dwarfing of the treated seedlings was noted although new growth appeared to be initiated normally.

In other experiments (Todd et al., 1956), avocado seedlings were fumigated for six months with ozonated gasoline vapour of sufficient strength to injure spinach, sugar beet, and tomato in one day. The fumigated seedlings showed no visible lesions but they had 7% less elongation of stems and 23% less increase in stem diameter than the controls.

(7) Kentia palms, which had been produced in Los Angeles, were exposed for eight months at Riverside (Todd et al., 1956). Some were grown in a lathhouse, some in an unfiltered greenhouse and some in a greenhouse supplied with carbon-filtered air. In the lathhouse only slight growth was made and the leaves were small and chlorotic. There was a little more growth and less chlorosis in the unfiltered greenhouse, but in carbon-filtered air the plants grew noticeably larger and had one extra leaf. The leaves were dark green in colour, and definitely longer than those in polluted air.

(8) Various spray and dust applications to vegetation have been experimented with to prevent smog damage (Kendrick et al., 1954; Middleton, 1956). Dithio- and bisdithiocarbamates in various commercial forms, including the zinc salt, and tetramethyl thiuram disulfide offered a considerable degree of protection to pinto beans provided the lower surface of the leaves was well covered. The concentration of the active material should be at least 10 % in the dust and 2 % in the spray. Protection falls to zero in about 16 days. Bordeaux spray gives no protection to beans.

The interesting observation has been made that a rust infection on pinto bean protects the leaf against smog injury (Yarwood & Middleton, 1954). Five times as much smog was required to injure the infected as the uninfected areas. Evidently the protecting substances diffuse beyond the mycelium of the rust. This protection remains after the rust itself is killed by dipping the leaf in water at 45° C for 90 seconds.

Other Pollutants

There are many other pollutants that are phytotoxic, but their discharge into the air in plant-damaging concentrations is extremely unlikely, except, perhaps, accidentally. These pollutants are as follows:

(1) Chlorine is about three times as phytotoxic as sulfur dioxide Thornton & Setterstrom, 1940; Zimmerman, 1949). Fortunately, its rooduction and use is confined largely to the chemical industry and it is (do valuable to waste. Limited plant damage is recorded due to gas from ta heavily chlorinated swimming pool in New York and a sewage disposal plant in California (Stout, 1932).

(2) Hydrogen chloride, of which about 10 p.p.m. for a few hours is required to cause incipient plant damage, was an important pollutant a century ago in the neighbourhood of the Le Blanc soda factories in England and Germany. When recovery of the gas as hydrochloric acid was instituted, the problem was solved and has had no serious import since that time.

(3) Ammonia has about the same phytotoxity as hydrogen chloride, in both cases perhaps the result of simple pH effects (Thornton & Setterstrom, 1940). Definite injury was observed on buckwheat, coleus, sunflower, and tomato with 40 p.p.m. for one hour; and slight marginal injury with 16.6 p.p.m. for four hours. The lesions had a "cooked green" appearance that became brown on drying.

(4) Hydrogen sulfide is only slightly phytotoxic (McCallan, Hartzell & Wilcoxon, 1936; Thornton & Setterstrom, 1940). Fumigation at 20-40 p.p.m. for five hours caused slight injury on a few species, but some could withstand 400 p.p.m. The most sensitive were cosmos, radish, clover,

PLATE 4

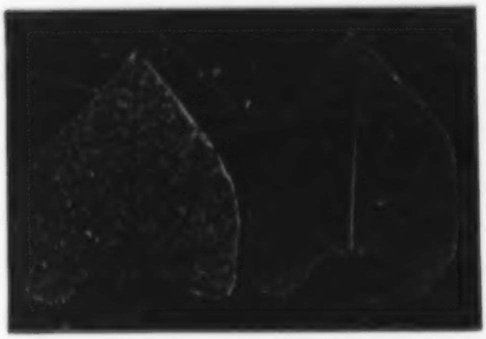


Fig. 15. Upper and lower surfaces of pinto bean treated with ozone (0.3 p.p.m., 2 hours)

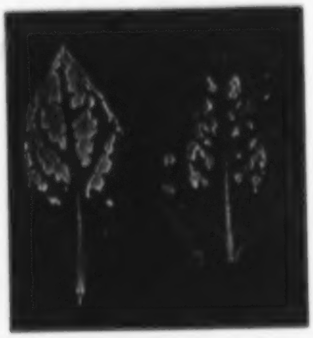


Fig. 16. Upper and lower surfaces of petunia treated with ozone (0.18 p.p.m., 6 hours)

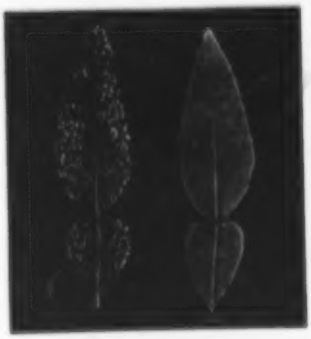


Fig. 17. Upper and lower surfaces of citrus injured by smog

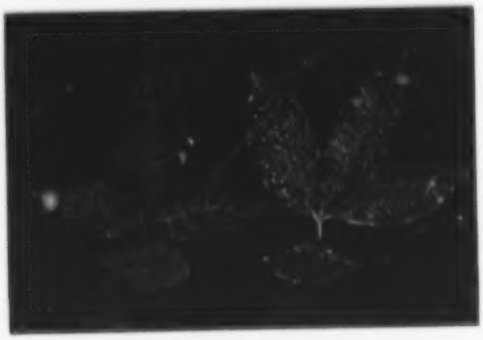


Fig. 18. White sapota damaged by ozone and smog

ig. 19. Ozone stipple on avocado (0.18 p.p.m., 7 days)

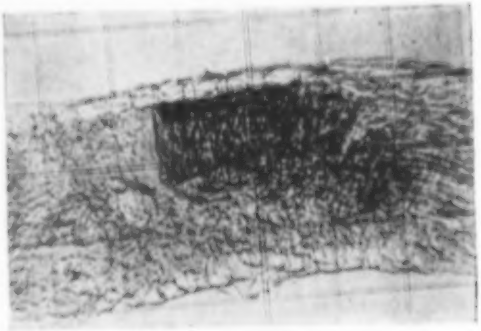


Fig. 20. Transverse section of grape leaf showing ozone injury on palisade cells (0.6 p.p.m 22.25 hours, at night)

Acknowledgement is made on page 275 to the persons who furnished the above coloured photographs.

tomato, poppy, salvia, cucumber, soybean, and aster; the intermediate in sensitivity were cornflower, buckwheat, nasturtium, sunflower, gladiolus, castor bean, and pepper; the resistant were coleus, peach, strawberry, cherry, apple, purslane, and carnation. Hydrogen sulfide near oil refineries, however obnoxious to the sense of smell, is probably harmless to plants. Bleasdale (1952b) found that the gas from a sewage disposal plant near Manchester counteracted to some extent the harmful effects of the city smoke on his rye grass.

(5) Hydrogen cyanide is sometimes present in manufactured gas to the extent of 200-300 p.p.m. (Hitchcock et al., 1934; Thornton & Setterstrom, 1940). It has injured the roots of plants in greenhouses when the gas pipes leaked below ground level. When citrus trees were covered with a tent and fumigated with hydrogen cyanide at concentrations up to 1100 p.p.m. for 40 minutes, the treatment sometimes injured the tree (Bartholomew, Sinclair & Lindgren, 1942). Injury in the open atmosphere is very improbable.

(6) Mercury is phytotoxic in greenhouses but not in the open atmosphere (Zimmerman & Crocker, 1934a).

(7) The new herbicides, such as 2-4 dichlorophenoxyacetic acid (2-4 D), affect some species but not others (Dunlap, 1948; Johnson, 1947; Middleton et al., 1956; Staten, 1947). Cotton, grapes and tomatoes are especially sensitive. In Texas, 10 000 acres (about 4000 hectares) of cotton were injured by 2-4 D at a distance of 15-20 miles (about 25-30 km) from rice fields that had been sprayed from aeroplanes. It is estimated that 1 g of 2-4 D could mark all the cotton plants on 25 acres (about 10 hectares) and 15 g could injure them permanently (Middleton et al., 1956).

Symptoms on cotton are: (*a*) leaves with scalloped margins, narrow lamina and long tentacle-like teeth; (*b*) stems swollen, with the cortex broken open, and spherical gall-like swellings near the ground; (*c*) squares and bolls malformed with reduced number of locules in the bolls; (*d*) new growth by lateral branching; (*e*) reduced germination of seed; and (*f*) retarded emergence of the hypocotyls (Dunlap, 1948). Somewhat similar symptoms were noted on roses, phlox, zinnia, petunia, tomato, grapes, pepper, tobacco, and cabbage.

As a result of the replacement of the volatile esters with non-volatile compounds, and after an intensive educational campaign to teach proper methods of handling and applying these compounds, plant injury due to their use has been greatly reduced in recent years in California (Middleton et al., 1956).

Summary

The literature on the effects of air pollution on plants has been reviewed with special reference to those pollutants that present major problems —viz., sulfur dioxide, hydrogen fluoride, London type smog, and Los

Angeles type smog. The others, which are definitely of minor importance, are referred to more briefly.

Sulfur dioxide has long been recognized as an air pollutant because it arises from the combustion of nearly all fuels, especially coal, and from the roasting of sulfide ores. It is phytotoxic in concentrations above 0.1 × 0.2 p.p.m., depending on the length of exposure. Below about 0.4 p.p.m., the gas tends to be oxidized in the cells as rapidly as it is absorbed, and interference with functions such as photosynthesis is slight. Toxic concentrations of sulfate are finally accumulated. Chronic rather than acute injury, if any, is generally manifested with these small concentrations. Above about 0.4 p.p.m., acute injury occurs more frequently, owing to the reducing properties of sulfite in the cells. Temporary interference with photosynthesis or "invisible injury" can occur to some extent, but these concentrations cause acute injury if maintained for more than short periods, and recovery is rapid when the fumigation is stopped. Practical methods of evaluating economic injury to vegetation have been summarized.

Hydrogen fluoride behaves somewhat similarly to sulfur dioxide, except that with a few species of plants it is effective in causing lesions and interfering with photosynthesis in concentrations two or three orders of magnitude smaller than in the case of sulfur dioxide. With most species it is up to ten times as effective as sulfur dioxide. Fluoride accumulated in the cells in sublethal amounts interferes with photosynthesis as does sulfite, but whereas the latter is deactivated by simple oxidation to sulfate, the former must be removed by translocation, volatilization, or some obscure chemical reaction, which makes much slower the recovery of the plant functions after hydrogen fluoride fumigation. There appears to be a concentration of hydrogen fluoride for each species below which "invisible injury" does not occur—ranging from a few parts per thousand million for the most sensitive to over 500 p.p.t.m. for the resistant plants. Forage may be rendered unsafe for animal feeding if more than 50 p.p.m. (dry basis) fluorine is absorbed.

London type smog may present essentially a sulfur dioxide problem, but the gaseous constituents as well as the aerosols need further evaluation.

The Los Angeles type smog is now fairly well understood as to its mode of formation and its phytotoxic effects, but the actual compounds that cause these effects are still unknown. The smog causes characteristic leaf lesions which are quite different from those produced by other pollutants, including ozone, which may be a constituent of the smog. It also causes some "invisible" injury. Visible damage to crops in Southern and Northern California was estimated at over US $85 000 000 and US $1 100 000 respectively, annually, in 1956.

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REFERENCES

Adams, D. F., Applegate, H. G. & Hendrix, J. W. (1957) *J. agric. Food Chem.*, **5**, 108

Adams, D. F., Mayhew, D. J., Gnagy, R. N., Richey, E. P., Koppe, R. K. & Allen, I. W. (1952) *Industr. Engng Chem.*, **44**, 1356

Adams, D. F., Shaw, C. G., Gnagy, R. N., Koppe, R. K., Mayhew, D. J. & Yerkes, W. D., jr (1956) *J. agric. Food Chem.*, **4**, 64

Agate, J. N., Bell, G. H., Boddie, G. F., Bowler, R. G., Buckell, M., Cheeseman, E. A., Douglas, T. H. J., Druett, H. A., Garrad, J., Hunter, D., Perry, K. M. A., Richardson, J. D. & Weir, J. B. de V. (1949) *Industrial fluorosis ; a report...*, London (Medical Research Council Memorandum No. 22)

Allmendinger, D. F., Miller, V. L. & Johnson, F. (1950) *Proc. Amer. Soc. hort. Sci.*, **56**, 427

Anderson, H. W. (1956) *Diseases of fruit crops*, New York, p. 306

Bartholomew, E. T., Sinclair, W. B. & Lindgren, D. L. (1942) *Hilgardia*, **14**, 373

Benedict, H. M. & Breen, W. H. (1955) In: *Proceedings of the third national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 177

Benson, N. R. (1959) *Proc. Amer. Soc. hort. Sci.*, **74**, 184

Bleasdale, J. K. A. (1952a) *Nature (Lond.)*, **169**, 376

Bleasdale, J. K. A. (1952b) Thesis, Manchester

Bobrov, R. A. (1952a) *Phytopathology*, **42**, 558

Bobrov, R. A. (1952b) In: *Proceedings of the second national air pollution symposium*, Los Angeles (Stanford Research Institute), p.129

Bobrov, R. A. (1955) *Amer. J. Botany*, **42**, 467

Bredemann, G. & Radeloff, H. (1937a) *Angew. Botan.*, **19**, 172

Bredemann, G. & Radeloff, H. (1937b) *Phytopath. Z.*, **5**, 195

Brennan, E. G., Leone, I. A. & Daines, R. H. (1950) *Plant Physiol.*, **25**, 737

Brewer, R. F., McColloch, R. C. & Sutherland, P. H. (1957) *Proc. Amer. Soc. hort. Sci.*, **70**, 183

Brisley, H. R., Davis, C. R. & Booth, J. A. (1959) *Agron. J.*, **51**, 77

Brisley, H. R. & Jones, W. W. (1950) *Plant Physiol.*, **25**, 666

Cann, G. R., Noble, W. M. & Larson, G. P. (1954) *Air Repair*, **4**, 83

Churchill, H. V., Rowley, R. J. & Martin, L. N. (1948) *Analyt. Chem.*, **20**, 69

Crocker, W., Hitchcock, A. E. & Zimmerman, P. W. (1935) *Contr. Boyce Thompson Inst.*, **7**, 231

Crocker, W., Zimmerman, P. W. & Hitchcock, A. E. (1932) *Contr. Boyce Thompson Inst.*, **4**, 177

Daines, R. H., Leone, I. & Brennan, E. (1952) *The effect of fluorine on plants as determined by soil nutrition and fumigation studies.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 97

Dean, R. S. & Swain, R. E. (1944) *U.S. Bur. Mines Bull.*, No. 453

De Ong, E. R. (1946) *Phytopathology*, **36**, 469

Dorries, W. (1932) *Z. Pflanzenkrankh. u. Gallenkinde*, **42**, 257

Dunlap, A. A. (1948) *Phytopathology*, **38**, 638

Erickson, L. C. & Wedding, R. T. (1956) *Amer. J. Botany*, **43**, 32

Glater, R. A. B. (1956) *Phytopathology*, **46**, 696

Griffin, S. W. & Bayles, B. B. (1952) *Some effects of fluorine fumes on vegetation.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 106

Haagen-Smit, A. J. (1952) *Industr. Engng Chem.*, **44**, 1342

Haagen-Smit, A. J., Darley, E. F., Zaitlin, M., Hull, H. & Noble, W. (1952) *Plant Physiol.*, **27**, 18

Haagen-Smit, A. J. & Fox, M. M. (1956) *Industr. Engng Chem.*, **48**, 1484

Hammett, F. S. (1930) *Proc. Amer. Philosoph. Soc.*, **69**, 217

Haselhoff, E., Bredemann, G. & Haselhoff, W. (1932) *Entstehung, Erkennung und Beurteilung von Rauchschäden*, Berlin

Haselhoff, E. & Lindau, G. (1903) *Die Beschädigung der Vegetation durch Rauch*, Leipzig

Heggestad, H. E. & Middleton, J. J. (1959) *Science*, **129**, 208

Hill, G. R., jr & Thomas, M. D. (1933) *Plant Physiol.*, **8**, 223

Hitchcock, A. E., Crocker, W. & Zimmerman, P. W. (1932) *Contr. Boyce Thompson Inst.*, **4**, 155

Hitchcock, A. E., Crocker, W. & Zimmerman, P. W. (1934) *Contr. Boyce Thompson Inst.*, **6**, 1

Holmes, J. A., Franklin, E. C. & Gould, R. A. (1915) *U.S. Bur. Mines Bull.*, No. 98

Hull, H. M. & Went, F. W. (1952) In: *Proceedings of the second national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 122

Johnson, E. M. (1947) *Phytopathology*, **37**, 367

Johnson, F., Allmendinger, D. F., Miller, V. L. & Gould, C. J. (1950) *Phytopathology*, **40**, 239

Katz, M. (1949) *Industr. Engng Chem.*, **41**, 2450

Katz, M. (1952) *The effect of sulfur dioxide on conifers.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 84

Kaudy, J. C., Brigham, F. T., McColloch, R. C., Liebig, G. F. & Vanselow, A. P. (1955) *Proc. Amer. Soc. hort. Sci.*, **65**, 121

Kehoe, R. A. (1949) In: *Proceedings of the first national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 115

Kendrick, J. B., jr, Darley, D. F., Middleton, J. T. & Paulus, A. O. (1956) *Calif. Agric.*, **10**, No. 8

Kendrick, J. B., jr, Middleton, J. T. & Darley, D. F. (1954) *Phytopathology*, **44**, 494

Koritz, H. G. & Went, F. W. (1953) *Plant Physiol.*, **28**, 50

Kotte, W. (1929) *NachrBl. dtsch. PflSchDienst (Berl.)*, **9**, 91

Largent, E. J. (1949) In: *Proceedings of the first national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 129

Leone, I. A., Brennan, E. G., Daines, R. H., & Robbins, W. R. (1948) *Soil Sci.*, **66**, 259

Loftfield, J. V. G. (1921) *Behavior of stomata*, Washington (Carnegie Institution of Washington Publication No. 314)

Los Angeles County Air Pollution Control District (1951) *Second technical and administrative report on air pollution control in Los Angeles County*, Los Angeles

McCallan, S. E. A., Hartzell, A. & Wilcoxon, F. (1936) *Contr. Boyce Thompson Inst.*, **8**, 189

McCombie, H. & Saunders, B. C. (1946) *Nature (Lond.)*, **157**, 776

MacIntire, W. H. et al. (1949) *Industr. Engng Chem.*, **41**, 2466

Massey, L. M. (1952) *Similarities between disease symptoms and chemically rinduced injus to plants.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United Statey Technical Conference on...*, New York, p. 48

Metcalfe, C. R. (1953) *Nature (Lond.)*, **172**, 659

Middleton, J. T. (1956) *J. Air Poll. Control Ass.*, **6**, 1

Middleton, J. T., Crafts, A. S., Brewer, R. F. & Taylor, O. C. (1956) *Calif. Agric.*, **10**, No. 6, 9

Middleton, J. T., Darley, D. F. & Brewer, R. F. (1957) *Damage to vegetation from polluted atmospheres*, Philadelphia (Proc. Amer. Petrol. Inst., Sect. III, Refining), 8 pp.

Middleton, J. T., Kendrick, J. B., jr & Darley, D. F. (1955) In: *Proceedings of the third national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 191

Middleton, J. T., Kendrick, J. B., jr & Schwalm, H. W. (1950a) *Calif. Agric.*, **4**, No. 11, 7

Middleton, J. T., Kendrick, J. B., jr & Schwalm, H. W. (1950b) *Plant Dis. Reptr*, **34**, 245

Miller, V. L. et al. (1952) *The effect of atmospheric fluoride on Washington agriculture*. In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 116

Miller, V. L., Johnson, F. & Allmendinger, D. F. (1948) *Phytopathology*, **38**, 30

National Research Council of Canada (1939) *Effect of sulfur dioxide on vegetation*, Ottawa (Publication 815)

Nichol, A. A., Benedict, H. M., Byrne, J. L. & McCarty, C. P. (1957) *J. Air Poll. Control Ass.*, **6**, 239

Nielsen, J. P., Benedict, H. M. & Holloman, A. J. (1954) *Science*, **120**, 182

Nielsen, J. P. & Dangerfield, A. D. (1955) *A.M.A. Arch. industr. Hyg.*, **11**, 61

Noack, K. (1929) *Z. angew. Chem.*, **42**, 123

Noble, W. M. (1955) *J. agric. Food Chem.*, **3**, 330

O'Gara, P. J. (1922) *Industr. Engng Chem.*, **14**, 744

Peters, R. A., Wakelin, R. W. & Buffa, P. (1953) *Proc. roy. Soc. B*, **140**, 497

Prince, A. L., Bear, F. E., Brennan, E. G., Leone, I. A. & Daines, R. H. (1949) *Soil Sci.*, **67**, 269

Radeloff, H. (1938) *Jber. hamburg. Inst. angew. Botan.*, **55**, 119

Radeloff, H. (1939) *Jber. hamburg. Inst. angew. Botan.*, **56**, 126

Remmert, L. F., Parks, T. D., Lawrence, A. M. & McBurney, E. H. (1953) *Analyt. Chem.*, **25**, 450

Rhine, J. B. (1924) *Botan. Gaz.*, **78**, 226

Richards, B. L., Middleton, J. T. & Hewett (1958) *Agron. J.*, **50**, 559

Rushton, A. G. (1921) *Ann. appl. Biol.*, **7**, 390

Scott, F. M. (1950) *Botan. Gaz.*, **111**, 378

Setterstrom, C. & Zimmerman, P. W. (1939) *Contr. Boyce Thompson Inst.*, **10**, 155

Setterstrom, C. Zimmerman, P. W. & Crocker, W. (1938) *Contr. Boyce Thompson Inst.*, **9**, 179

Smith, O. D. & Parks, T. D. (1955) *Analyt. Chem.*, **27**, 998

Solberg, R. A. & Adams, D. F. (1956) *Amer. J. Botany*, **43**, 755

Stanford Research Institute (1954) *The smog problem in Los Angeles County*, Los Angeles (Western Oil and Gas Association)

Staten, G. (1947) *J. Amer. Soc. Agron.*, **38**, 536

Stephens, E. R., Darley, E. F., Taylor, O. C. & Scott, W. E. (1960) *Proc. Amer. Petrol. Inst.* (in press)

Stephens, E. R., Hanst, P. L., Doerr, R. C. & Scott, W. E. (1956a) *Industr. Engng Chem.*, **48**, 1498

Stephens, E. R., Scott, W. E., Hanst, P. L. & Doerr, R. C. (1956b) *J. Air Poll. Control Ass.*, **6**, 159

Steward, F. C., Thompson, J. F., Millar. F. K., Thomas, M. D. & Hendricks, R. H. (1951) *Plant Physiol.*, **26**, 123

Stoklasa, J. (1923) *Die Beschädigung der Vegetation durch Rauchgase und Fabriksexhalation*, Berlin

Stout, G. L. (1932) *Calif. Dept. Agr. Monthly Bull.*, **21**, 340

Swain, R. E. & Johnson, A. B. (1936) *Industr. Engng Chem.*, **28**, 42

Taylor, O. C., Cardiff, E. A., Mersereau, J. D. & Middleton, J. T. (1957) *Calif. Agric.*, **11**, No. 3, 9

Taylor, O. C., Stephens, E. R., Darley, E. F. & Cardiff, E. A. (1960) *Amer. Soc. hort. Sci.*, **75** (in press)

Thomas, M. D. (1951) *Ann. Rev. Plant Physiol.*, **2**, 293

Thomas, M. D. (1956) *J. Air Poll. Control Ass.*, **5**, 1

Thomas, M. D., & Hendricks, R. H. (1956) *Effect of air pollution on plants.* In: Magill, P.L., Holden, F. R. & Ackley, C., ed. *Air pollution handbook*, New York, Section 9, p. 1

Thomas, M. D., Hendricks, R. H., Bryner, L. C. & Hill, G. R. (1944) *Plant Physiol.*, **19**, 227

Thomas, M. D., Hendricks, R. H., Collier, T. R. & Hill, G. R. (1943) *Plant Physiol.*, **18**, 345

Thomas, M. D., Hendricks, R. H. & Hill, G. R. (1944) *Plant Physiol.*, **19**, 212

Thomas, M. D., Hendricks, R. H. & Hill, G. R. (1950a) *Industr. Engng Chem.*, **42**, 2231

Thomas, M. D., Hendricks, R. H. & Hill, G. R. (1950b) *Soil Sci.*, **70**, 9

Thomas, M. D., Hendricks, R. H. & Hill, G. R. (1952) *Some impurities in the air and their effect on plants.* In: McCabe, L.C., ed. *Air pollution. Proceedings of the United States Technical Conference on...*, New York, p. 41

Thomas, M. D. & Hill, G. R. (1935) *Plant Physiol.*, **10**, 291

Thomas, M. D. & Hill, G. R. (1937) *Plant Physiol.*, **12**, 285

Thornton, N. C. & Setterstrom, C. (1940) *Contr. Boyce Thompson Inst.*, **11**, 343

Todd, G. W. (1955) *Analyt. Chem.*, **27**, 1490

Todd, G. W. (1956) *Physiologia Plantarum*, **9**, 421

Todd, G. W., Middleton, J. T. & Brewer, R. F. (1956) *Calif. Agric.*, **10**, No. 6, 7

Warburg, O. & Christian, W. (1941) *Naturwissenschaften*, **29**, 590

Wedding, R. T. & Erickson, L. C. (1955) *Amer. J. Botany*, **42**, 570

Went, F. W. (1955) In: *Prodeedings of the third national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 8

Wilkins, E. T. (1954) *Mech. Engr*, **76**, 426

Wislicenus, H. (1914) *Sammlung von Abhandlunger über Abgase und Rauchschäden*, Berlin

Yarwood, C. E. & Middleton, J. T. (1954) *Plant Physiol.*, **29**, 393

Zimmerman, P. W. (1949) In: *Proceedings of the first national air pollution symposium*, Los Angeles (Stanford Research Institute), p. 135

Zimmerman, P. W. (1955) *Professional Paper* 2, 124-145, Boyce Thompson Institute of Plant Research

Zimmerman, P. W. & Crocker, W. (1934a) *Contr. Boyce Thompson Inst.*, **6**, 167

Zimmerman, P. W. & Crocker, W. (1934b) *Contr. Boyce Thompson Inst.*, **6**, 455

Zimmerman, P. W. & Hitchcock, A. E. (1956) *Contr. Boyce Thompson Inst.*, **18**, 263

Zimmerman, P. W. Hitchcock, A. E. & Crocker, W. (1931) *Contr. Boyce Thompson Inst.*, **3**, 459

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ECONOMIC AND SOCIAL ASPECTS OF AIR POLLUTION

Introduction

In considering the vast subject of the consequences of air pollution, many aspects must be taken into account, among which are the following: (*a*) effects on human and animal health; (*b*) effects on normal plant growth; (*c*) degeneration of objects useful to man (through dirt, accentuated corrosion, etc.); (*d*) hindrance or even damage to industrial activities caused by certain emissions; (*e*) various nuisances due to intensified and sometimes noxious fog, and to mists which absorb the sun's rays, etc. All these effects are the subject of special research to determine the exact nature of the damage inflicted, its extent, mechanism and degree of intensity.

The purpose of this chapter is to study not the disadvantages themselves but their economic and social repercussions, in order to lead to a better appreciation of the bases and limits of the measures which must be adopted for the rational control of regional air pollution.

Thorough study of the extensive literature on air pollution leads, in our opinion, to the conclusion that the bases used for the measurement of progress in the control of this nuisance are sometimes too conventional and only indirectly related to the damage caused, which, nevertheless, has a real effect on, and is definitely felt by, the inhabitants of the affected areas.

Under a semblance of accuracy, often more attention is paid to qualitative than to quantitative aspects. It is true that among the existing quantitative data we find extensive—and valuable—documentation on the limit doses of gases, vapours, and dusts which may to a greater or less extent affect human and animal health or plant life. For the large towns and other built-up areas there are statistics showing the extent of the polluted zones, but the pollution is expressed in figures based on standard tests, such tests being selected according to their more or less adequate interpretation of a particular effect of pollution, i.e., directly measurable damage. In these circumstances, it is evident that the evaluation of a method for the combating of such pollution is a delicate matter which must therefore be conventionally defined. In fact, when an inhabitant of a polluted area reads a report, he often finds that the statistical results do not in any way tally with his own personal idea of the pollution from which he suffers. For example, the rate of reduction of the weight of the dust collected in some conventional apparatus is, for the layman, less eloquent than the possibility of reducing house maintenance costs. This is not, however, intended as a condemnation or criticism of the usual methods of assessment; our purpose is merely to emphasize the value of establishing a known correlation between the results of a standard test and the economic data, for example.

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Unfortunately, it is not possible immediately to translate all air pollution effects into economic terms. Odours are an example. An odour will constitute a subjective discomfort for a given person, but it is difficult to evaluate such discomfort statistically. At the most, the effect of a deodorant may be studied and an odour reduction rate established, indicating the extent to which the odour may be reduced (without completely eliminating it), so that it will not be noticed by the inhabitants of the area in question. A harmful smoke affecting the general health may be represented as a " loss " on the assumption that it increases costs for care and treatment. But it is not so easy to express the physical and psychological effects in terms of money.

It is obvious, therefore, that many different factors and conditions must be taken into account when considering the economic and social aspects of air pollution. We will here confine ourselves to the enumeration of these economic and social aspects, briefly commenting upon them and completing the list with the practical data which have already been collected and which provide examples capable of placing the problem in its true perspective.

Economic Aspects of Pollution

Evaluation of damage: bases for calculation

Except in special cases, damage is estimated by fairly simple, but justifiable, extrapolation—in the absence of any better method and in view of the complexity of the problem. The damage would, of course, be calculated on different bases according to: (*a*) the type of object damaged (plants, metal-work); (*b*) the type of pollution causing the damage; (*c*) other concomitant causes of damage; (*d*) local methods of assessing the degree of pollution; and (*e*) the quality of statistical data concerning the bases for these various estimates.

Before passing on to examples of calculation of loss expressed in money, we will consider briefly the difficulties inherent in this type of calculation. There is " total " damage: flowers destroyed or crops rendered unsalable are examples of " total " damage, and this can easily be calculated. The case is different if—as often happens—the damage imputable to pollution is only partial. In this case, a " reduction of value coefficient " must be adopted on the basis of the results of investigations into the relative rate of pollution.

This relative rate, when it is known, makes calculation of damage easy, but the difficulty resides in the choice of a test for the assessment of pollution. Should this be the local dust rate, the local SO_2 rate, the light reduction rate, etc.? These are all questions to be debated in relation to the type of damage. In the case of plants, the SO_2 rate may be taken as a basis for evaluation of the nuisance; in the case of corrosion, SO_2 may be a factor, but near the sea it will be necessary to take the chloride rate into account—and so on.

In our opinion, thorough study of these bases for calculation would be useful, and it is our intention in this chapter to commence by an examination of some of the studies already made, with a view to drawing lessons for the future.

The other aspect of the question is the evaluation of the cost of the measures adopted to combat or eliminate pollution, in order to have some idea of operational costs. In this instance, direct calculation is more feasible, in view of the fact that the costs of measures for the reduction of dust or for the purification of emissions are known; the number of installations provided, or to be provided, can also be established on the basis of governmental or private inquiries.

Examples of economic repercussions of pollution

The economic losses due to air pollution are as follows:

(1) Losses due to direct or indirect effects of pollution:

- (*a*) on human health;
- (*b*) on livestock;
- (*c*) on plants.

(2) Losses due to corrosion of various materials and of their protective covering.

(3) Losses due to maintenance costs, inside and outside houses and other buildings; to depreciation of objects or merchandise exposed to pollution (some of these losses are covered by paragraph 2 above).

(4) Losses due to unburnt residue in chimneys, motors, and various engines which contributes to the general pollution.

(5) Expenditure directely incurred by the adoption of technical measures for the suppression or reduction of smoke or emissions from factories.

(6) Increase in cost of electric power due to dust removal and other treatment of smoke from power or nuclear stations.

(7) Various losses due indirectly to pollution: increased transport costs in periods of smog; wasted electricity due to premature twilight caused by smoke, etc.

(8) Expenditure in connexion with the administrative organization of pollution control.

(9) Costs of medical, agricultural, chemical, and physico-chemical research aimed at working out techniques for the measurement of pollution; investigating the ways in which pollution affects persons, animals, plants, and materials; and studying appliances or installations for the reduction or suppression of various dusts and emanations.

It is difficult to find in the literature any statistical data relating to economic effects. We will therefore group together certain types of damage which are estimated *en bloc* in studies which we have been able to consult. In order that these data may be generalized and so that future information may be improved and completed, the bases for the statistics and the correlation between damage and the test for the measurement of air pollution will be indicated where possible.

Damage to living creatures (health aspect)

Reference has already been made to some of the difficulties in assessing the direct effect on living creatures. Table 1 gives some French statistics showing the differences in numbers of deaths from respiratory diseases as between urban and rural areas.

Data of this type have been collected in a number of American towns, including Detroit-Windsor, where a Joint International Commission has set up a Technical Advisory Board on Air Pollution, and Cincinnati (Kettering Laboratory Survey) (Organisation for European Economic Co-operation, 1957). In these studies, an effort is made to establish a relationship between the state of health of the population and the results of measurement of pollution by American techniques.

It is impossible to obtain a clear picture (except in rare cases such as localized accidents) of the extent of medical services made necessary by air pollution. At the most, it may be assumed that such pollution is one of the determining factors in the appearance of certain diseases. Part of the social security budget is devoted to covering the costs involved, but their distribution by cause of disease is extremely difficult.

In order to complete the picture, an attempt should be made to estimate the effect of pollution on human output. Such output depends to some extent on the regularity and punctuality of transport, which are impaired by smog. The effect of pollution on the health of persons living in affected districts also plays a part in reducing output.

TABLE 1

NUMBERS OF DEATHS FROM RESPIRATORY DISEASES IN URBAN AND RURAL AREAS (PER 100 000 INHABITANTS)

Cause of death	Urban populations of			Rural populations
	100 000 inhabitants and over	50 000-100 000 inhabitants	Under 50 000 inhabitants	
Pneumonia	47.90	39.22	35.75	31.55
Bronchitis	61.56	53.82	48.77	36.94
Other respiratory disorders (with the exception of influenza)	11.19	9.71	10.60	9.66
Total	120.65	102.75	95.12	78.15

Source: Pannetier (1957).

General material damage

USA

In 1911-13 the Mellon Institute undertook an important economic study of nuisances caused by smoke (McCabe, 1949). The estimates made at that time (1913), relating to Pittsburgh, were as follows:

(*a*) US $2 000 000 were spent in the supplementary washing and dry-cleaning of clothes;

(*b*) US $750 000-1 000 000 were spent on the maintenance or renewal of wallpapers and curtains;

(*c*) US $2 000 000 were spent on measures for the special protection of merchandise or on the replacement of merchandise made valueless by smoke;

(*d*) For the year in question, the total losses amounted to US $8 500 000.

In 1950-51 it was estimated that the total losses due to pollution in the whole of the USA amounted to US $1 500 000 000, i.e., US $10 per inhabitant per year (Johnson, 1952). A considerable part of these losses referred to crop damage. In 1950, losses in the Los Angeles region with respect to certain food crops amounted to US $300 000. Referring to these figures, Johnson suggests that the measures for the control of pollution undertaken by the Government, the local authorities and industry cost about US $100 000 000 per year.

In 1937, a survey was made in Chicago and St Louis, on the basis of which it was estimated that the annual losses due to smoke in the two cities were US $30 000 000 and US $19 000 000 respectively (McCabe, 1949).

The foundries of the Consolidated Mining and Smelting Company at Trail caused SO_2 pollution of the Columbia River Valley during the first year of operation, and the Company was ordered to pay US $350 000 compensation for the resulting damage to crops. Gibson (1949) estimates that the annual losses in the region are in the neighbourhood of US $500 000 000. In this connexion, it should be emphasized that the expenditure incurred in combating noxious emissions at the source amounted in 1948 to US $100 000 000.

It is calculated that damage amounting to US $3 000 000 has been done to crops in the Los Angeles region every year since 1953, and this covers only visible damage.

Great Britain

Some estimates of the cost of pollution in Great Britain are as follows (Meetham, 1952):

Manchester, 1919	£1 per inhabitant per year
London, 1924	£1 4s. per inhabitant per year
For Great Britain as a whole:	
1924	£40 000 000-50 000 000 per year
1947	£100 000 000 per year

Assuming that 50 % of damage is due to smoke, it is calculated that the 2 400 000 tons[1] of smoke emitted per year causes damage to the extent of £50 000 000, i.e. one ton of smoke causes damage to the value of £20. Bearing in mind that smoke represents 1.33 % of the coal consumed, it is estimated that one ton of coal consumed causes damage to the value of five shillings.

Another detail in connexion with the United Kingdom is that, according to area, the external maintenance of shops must be undertaken at the following intervals:

Rural areas	every three years
Moderately polluted areas	every two years
Very polluted areas	every year

The costs incurred for cleaning and painting buildings is 15%-40% higher in polluted towns than in smokeless districts.

France

From a study made in France (Pannetier, 1957), the following details emerge:

	French francs
Supplementary expenditure on cleaning and provision of clothing	60 000 000 000
Painting and maintenance	15 000 000 000
Maintenance of buildings	60 000 000 000
Reduction of earnings	40 000 000 000
Miscellaneous: medical services	65 000 000 000
	240 000 000 000

i.e., Fr. fr. 6000 per inhabitant per year.

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[1] 1 long ton = 1.016 metric tons.

The French statistics provide the following information. The city dweller gives on an average 3 kg of clothing to be washed per week, as against a little over 2 kg for the rural inhabitant. This means 50 kg more per year which the town dweller has washed, or washes himself, and if it is admitted that this represents on an average Fr. fr. 200 per kg, it follows that the town dweller spends Fr. fr. 10 000 more than the rural inhabitant on keeping clothes at the same degree of cleanliness. One-fifth of this sum is undoubtedly spent on account of the particularly heavy dust content of the air in the towns. Assuming that 20 million of the inhabitants of France live in polluted zones, this extra cost amounts to Fr. fr. 40 000 000 000. In addition, it may be estimated that the atmosphere in urban agglomerations decreases the life of garments by one-twentieth and that, in France, urban dwellers account for half of the annual garment consumption (800 000 000 000), so that air pollution costs another Fr. fr. 20 000 000 000.

More frequent maintenance and renewal of paintwork. Similarly, taking as a starting point the Fr. fr. 100 000 000 000 annual turnover in the house-painting and wallpaper trade, and assuming that half of this amount refers to the urban areas and that a coat of paint or paper lasts six years in the country and four years in the towns (figures arrived at by building-trade experts), air pollution causes supplementary expenditure for repainting or repapering to the extent of Fr. fr. 100 000 000 000 every six years, i.e., about Fr. fr. 15 000 000 000 per year. It would seem justifiable, moreover, to double this figure, for other repair work is usually necessary in addition to papering and painting, particularly in the maintenance of shops and commercial establishments.

Maintenance of buildings. The depreciation of the zinc-work on roofs and the soiling of façades constitute the main damage caused to buildings by air pollution. Fuel oil, now being used more and more for heating, contains on an average 2 %-3 % of sulfur, which, after combustion, remains in the form of SO_2 or SO_3. These anhydrides are easily absorbed by the soot which forms in chimneys, and when this is made more or less damp by the weather and is deposited on the zinc-work of a roof, it eats into the metal to a considerable extent. Builders say that zinc-work, which used to last for 30 years, now has to be renewed in 4-5 years in the worst cases. Calculating 40 persons to a building, half in the urban areas, with zinc-work representing approximately Fr. fr. 200 000, this makes a further wastage of Fr. fr. 20 000 000 000 on account of air pollution. About twice this amount has to be spent on maintenance of external paintwork, various types of refacing, etc., which means an average waste of Fr. fr. 60 000 000 000. And in this figure no allowance is made for the more rapid depreciation, with the years, of buildings in large towns as compared with those in less congested areas.

Special studies relating to atmospheric corrosion

Corrosion Commissions convened by the Society of Industrial Chemistry in Brussels have made a study of the effect of pollution on metal-work. The intensity of the action varies according to the material; it also varies with the type of pollution, and the process may develop at varying rates. It may be asked to what extent the corrosion caused by average industrial pollution is greater than that occurring in a normal atmosphere or in sea air. Table 2 gives a few figures.

TABLE 2

CORROSION OF IRON AND ZINC IN DIFFERENT AREAS

Area	Relative annual penetration (1/1000")	
	Fe	Zn
Tropical	0.1	0.02
Rural	2.5	0.12
Marine	3.1	0.14
Industrial	5.4	0.62

Source: Gilbert (1954).

TABLE 3

DURABILITY OF STEEL IN RELATION TO CONDUCTIVITY OF RAINWATER

Station	Relative conductivity in mhos x 10^{-4}	Increased durability in relation to very polluted atmosphere
Marley	4 000	1
Ruysbroek	2 000	1.2
Liège	1 000	1.5
Mol	250	2.8

Source: Bermane (1953).

In Belgium, the durability of steel with two coats of minium, iron oxide and linseed oil has been related to the conductivity of rainwater, as shown in Table 3.

The data in Table 4 show the durability of steel in different types of atmosphere.

TABLE 4

DURABILITY OF STEEL IN DIFFERENT ATMOSPHERES

Steel constructions		Life in years		
Metal facing	1/1000"	Pittsburgh	State College	Key West
		Industrial	Rural	Marine
Zn	4.3	7	> 23	> 22
Al	4.0	> 10	> 23	> 22

Source: Gilbert (1954).

A correlation has been established between atmospheric pollution and the corrosion of iron (Table 5), using as the pollution test the weight of solid matter deposited on the surface unit.

TABLE 5

CORROSION OF IRON IN RELATION TO DEPOSIT ON SURFACE UNIT

	Rural atmosphere	Marine atmosphere	Industrial atmosphere	
			Woolwich	Sheffield
Deposit	3.4	7.8	11.7	22.3
Corrosion 1/1000"/year	0.9	1.1	1.8	4

Source: Hudson (1948).

Loss of materials in emissions

In 1926 the Mellon Institute estimated that 160 000 short tons [1] of nitrogen were lost in the smoke from coal used in domestic installations, i.e., the equivalent of half the consumption of nitrogen in the USA (Doyle, 1953). According to the US Bureau of Mines, 700 000 tons of manganese are lost every year in emissions, again representing 50 % of the country's manganese requirements (Doyle, 1953). In Great Britain, coal cinders pour into the air every year 1000 tons of gallium and 2000 tons of germanium. It is not, of course, possible to recover all these products economically.

Substantial losses are also due to unburnt residue in many fuels. The British statistics show that 1.3 % of the coal used is lost in smoke, which gives an indication of the average for a highly industrialized country: the annual 24 000 000 tons of smoke represent a loss of £6 000 000. If the invisible unburnt residues are taken into account, the losses in Great Britain amount to £20 000 000. The losses with respect to domestic fires are considerable, varying according to type of fire and type of fuel. In 1951, S. R. Craxford, of the Fuel Research Station, stated that in Great Britain the burning of coke and coal caused the emission of 2 300 000 tons of sulfur. At the same time, the country was importing 380 000 tons of sulfur from the USA. In 1953, 300 000 tons were imported against a consumption of 350 000 tons.

Economically speaking, the situation was as follows. If all the sulfur from coal (1 %) were recovered, one unit of such sulfur would cost 10 shillings, whereas this unit is bought by the acid factories for 2.5 shillings. According to tests in a pilot station, where the recovery process consists in the washing of smoke from power stations, it was found that the cost of treatment would be 8-9 shillings per ton of coal burnt. Parker nevertheless pointed out in 1954 that with the new techniques, the cost of purification would be brought down to 2 shillings per ton of coal with 1.5 % sulfur (85 % efficiency) (Parker, 1954).

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[1] 1 short ton = 0.9 metric ton.

The unburnt residue losses with respect to motor-cars in towns are very high, varying according to type of engine, power of engine, traffic conditions, etc. Los Angeles statistics give 7 % as the proportion of unburnt residue in emissions from cars in that town.

In assessing pollution costs, the expenditure on purification cannot always be considered as a total loss; it is sometimes possible to recover materials. There is, for example, the recovery of copper from foundry gases in the USA which has brought in US $27 000 000 (Keyser & Munger, 1952).

Costs of purification and control of pollution

In the county of Los Angeles, the cost of pollution control is 27.75 cents per capita per year, i.e., the quite insignificant amount of the price of a gallon of petrol (Los Angeles Air Pollution Control District, 1955). Nevertheless, the annual cost to industry in the period 1949-55 was US $9 000 000. The measures adopted from 1947 to 1955 cost US $35 000 000.

The average expenditure in the towns in the USA is something like 7 cents per capita per year, while industries and local and governmental authorities make an outlay of US $100 000 000 per year for the control of damage done by pollution. Added to this, there is the fact that the measures adopted to combat the emission of these vast quantities of waste matter into the air entail large-scale stoppages which industrial undertakings are obliged to take more and more into account in their general economy (Larson, 1954). In a power station, the installation costs for an electrofilter vary from US $1.5 to US $5 per cu.ft/min., according to the size of the installation. In large modern power stations, therefore, such purification measures will involve heavy expenditure. At the Battersea Power Station, London, about £2 000 000 were spent on attempts to eliminate SO_2.

Here we would mention also that to the already very high cost of installations for the removal of dust or for the purification of gases must sometimes be added expenditure on other important installations. In the case of removal of dust by a wet process, for example, means must be devised for the economic evacuation of the waste water, and in view of the regulations with regard to water-courses, the factory may be obliged to provide large water-treatment installations for the collection of sludge. This sludge must necessarily be dumped somewhere, and it in turn constitutes a new source of dust. Thus the solution of the smoke problem is often complicated by other important economic problems.

Social Aspects of Pollution

Socially speaking, air pollution is a factor which undermines the well-being of affected populations. It is inherent in all congested areas, and in all industrial undertakings unless these latter are provided with every possible facility for effectively combating the emissions. Industrial development has certainly increased the risks, but at the same time has led to the perfecting of processes for the prevention of the nuisance, so that in areas which were formerly highly polluted—Pittsburgh, for example—the quality of the air is now constantly improving, in a regular and spectacular manner.

In the large centres at least, the emissions from individual domestic fires have considerably decreased and furthermore greater comfort has been provided by the substitution of central heating. On the other hand, the pollution which was formerly caused by individual fires has now been replaced by that due to fuel oil. In addition, the enormous growth of motor traffic means that there is another source of pollution which is continually on the increase, caused by the emanations from vehicles, by the dust they raise and by the increased servicing and stocking in the larger centres to meet the needs of an ever-expanding clientèle. Moreover, in many places these vehicles running on petrol, or—more serious—on fuel oil, replace the electric trams which, of course, caused no pollution. On the other hand, the suppression of steam trains has changed the situation along the railroads. The substitution of the electric train has solved the problem of pollution, although at the moment a great deal of attention is being paid to the question of Diesel engines. Traffic in ports, on rivers and canals also raises problems: the use of craft running on petrol instead of coal or similar fuel has done away with the dense smoke, but it has not solved all the problems.

The social aspect of pollution may also be considered from the point of view of the future. What lies ahead? Industry is certainly expanding rapidly, and regions which were formerly free from pollution are now becoming exposed. In our view, those responsible for this expansion should include in their preliminary studies the question of the air pollution to which the inhabitants living near the industry will be subjected. Such studies should cover not only the effects on the health of the inhabitants, but also the effect on crops. The choice of site, taking into account the topographical and meteorological conditions, is extremely important if repetition of disasters and disquieting social problems such as those of the Meuse Valley and Donora are to be avoided.

The medical aspect of these problems is very complex. Statistics are unfortunately scarce, but their compilation would be highly desirable in the interests of devising suitable precautions.

The general public should be informed of the situation, not only so that they may understand it and appreciate the extent of the efforts made for their benefit, but also so that they may become aware of their own responsibilities in this matter of air pollution.

Physical and chemical studies are no doubt continuing, but substantial encouragement should be given in the search for a relationship between the scientific data and the health of the community, for otherwise extremely expensive measures may be adopted without there being any guarantee that they will improve general living conditions in the regions in question.

The systems for the measurement of pollution are multiplying. Interest is very great in devices permitting continuous measurement, for this is the only way of detecting the occasional emission, over short periods, of immense quantities of polluting material—which is the real trouble in some areas. People without special knowledge of the subject find it difficult to understand that it is not easy to detect the clandestine emissions which annoy them. Real difficulty is nevertheless encountered with many of the standard instruments, which give only the average pollution over fairly long periods.

Legislation for the protection of communities will have to be flexible in order to cover the differing local and operational conditions, but it should also prescribe measures which can be easily applied, such as the prohibition of certain fuels and certain types of unsatisfactory appliance.

The community is benefiting today from the great progress made in purification techniques and in the design of appliances, whether for the treatment itself or for control. There is no doubt that this progress—the result of discoveries in other fields—has led to the solution of many problems which were formerly insoluble. There are, nevertheless, some difficulties for which no satisfactory remedy has as yet been found, and among these is that of the emission of low concentration sulfurous smoke. The odour problem, which in many cases has been solved, still worries the responsible authorities.

In general, therefore, it may be said that the rise in the number of sources of pollution coincides with constant progress in methods of combating this nuisance. In the future, therefore, if progress continues, there is no reason to expect that living conditions will become more uncomfortable. Nevertheless, the public must be given at least elementary information on these problems and the financial means and techniques for overcoming them, so that the progress made may be appreciated. In addition, those whose task it is to study the problems and to adopt the necessary measures should be given adequate support, help and encouragement.

REFERENCES

Bermane, M. D. (1953) *Bull. Ass. belge Et. Mat.*, No. 7

Doyle, H. N. (1953) *Publ. Hlth Rep. (Wash.)*, **68,** 858

Gibson, W. B. (1949) *The economics of air pollution.* In: *Proceedings of the first national air pollution symposium,* Los Angeles (Stanford Research Institute), p. 109

Gilbert, P. T. (1954) *Ind. chim. belge,* **19,** 923

Hudson, J. C. (1948) *Ossature métallique*, No. 11, 239

Johnson, V. W. (1952) *Air pollution in relation to economics.* In: McCabe, L. C., ed. *Air pollution. Proceedings of the United States Technical Conference on . . .*, Washington, p. 37

Keyser, N. H. & Munger, H. P. (1952) In: *Symposium on air pollution*, Chicago (American Foundrymen's Society)

Larson, G. P. (1954) (Los Angeles County Air Pollution Control District, Publication No. 58)

Los Angeles County Air Pollution District (1955) *Annual report, 1954-1955*, Los Angeles

McCabe, L. C. (1949) *Trans. industr. Hyg. Found. America, Bull.*, No. 13, 91

Meetham, A. R. (1952) *Atmospheric pollution : its origin and prevention*, London, p. 268

Pannetier, G. (1957) *Techn. sanit. munic.*, **52,** 73

Parker, A. (1954) *The social and industrial consequences of the presence of sulphur in fuels.* In: Institute of Fuel. *Special study of sulphur removal and recovery from fuels. Volume 1 : Papers for the opening conference*, London, p. 5

Organisation for European Economic Co-operation, European Productivity Agency (1957) *Air and water pollution : the position in Europe and in the United States*, Paris

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CONTROL OF AIR POLLUTION BY SITE SELECTION AND ZONING

Introduction

The planning and zoning process

Planning is the over-all consideration of the use to which land in a particular area might be put, appropriate weighting being given to the peculiarities of specific local sites. Zoning is the legally instrumented guide to private and public land use, preferably in accord with a comprehensive plan, although this is not mandatory in many jurisdictions (California State, 1955). American practice, for example, considers that the two processes may be undertaken separately (New York State Department of Commerce, 1953), while British practice regards them as indivisible.

Zoning is an exercise of police power (regulation based on public health, safety, welfare, and convenience) made available by appropriate legal process to the local jurisdiction in order to control the orderly use of buildings and land (Gallion & Eisner, 1950; Horack & Nolan, 1955). Most police powers, such as those enforcing health or building regulations, are applied uniformly throughout the local jurisdiction. Zoning is different in that it can " district " or regulate differently various portions of the jurisdiction but must be uniform within such smaller areas. The zoning jurisdiction may cover one or several combinations of local governmental units, such as city, county, area, district, region, or metropolitan area.

The objectives of zoning are manifold— e.g., to lessen traffic congestion, to minimize fire and other hazards, to provide adequate light and air, to prevent overcrowding of land, to facilitate the provision of transportation,

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utilities and other facilities. Traditional zoning effectively decentralizes or clusters uses in new areas has a limited effect on existing uses in old areas, and has the marked disadvantage of divided authority in metropolitan areas containing many cities. The leeside of one city may be the windward side for an adjacent city. For effective control, larger air pollution zoning jurisdictions or districts, or even inter-state compacts or international agreements, may be advisable depending on meteorological conditions. Although local governments are accepted as the level for air pollution control, some authors have advocated that the responsibility should not descend to the city level but be at the metropolitan level with jurisdiction over cities within the district (Association for Planning and Regional Reconstruction, 1950; Chute, 1955; *National Conference on Metropolitan Problems*, 1956; California University, Bureau of Public Administration, 1957). In passing, it might be noted that governmental control of air pollution is not entirely accepted by some segments of society on the grounds that control is a technological rather than a legal problem (MacDonald, 1956). Opposing viewpoints draw attention to the fact that successful smoke abatement occurred only after governmental control was applied, and that timely and effective air pollution abatement require such control (American Society of Planning Officials, 1955a; Hoover, 1948).

The role of zoning as a means of air pollution control has been a subject of frequent discussion by many control experts, who have generally placed primary emphasis on the control of the contaminants at the source. Yet, control devices are not available for all contaminants; and the control is not absolute even in those cases in which devices do exist. Air pollution zoning could be effective in controlling the effects of residual emission after partial control or to gain time in which to develop better source-control technology. Practices that are not themselves pollution sources sometimes generate sources of air pollution. For example, a clean or smog-free electronics assembly plant might induce smog as a result of the automotive operations incident to operation of the concern. General zoning and planning could be utilized to reduce the mobile smog source by decentralization of industry, encouragement of rapid transit, elimination of automobile traffic in congested city centres, use of the free-flow type of roadway or freeway, and reduction of home to work travel.

The nature of air pollution control

Discussion of means of combating area emission problems requires knowledge of existing control techniques. There are two opposing theories of aerial waste disposal—namely, concentrate and contain, or dilute and disperse (Lieberman, 1957).

The concept of concentration and containment is primarily that of the control of the individual pollution source by installation of a properly engineered collection device. This can be augmented by restricting the industrial sources to a single area wherein their emissions will not affect their neighbours. In some cases, the specific contaminant prevents efficient collection, and is more easily converted to an innocuous substance and dispersed; examples occur in odour abatement by incineration and catalytic combustion in solvent control. Sometimes it may be necessary to alter completely the process fuel or material in order to prevent uncontrollable pollution downstream from the source.

When we turn to the alternative technique of dilution and dispersion in space and time, there is a close analogy with the problem of water pollution control. In considerations of water pollution control the boundaries are the stream bed and water surface, which are to be compared, in the case of aerial dispersal, to terrain features plus the presence of an "inversion base" acting as the uppermost surface of the air flow. In some areas and at some times the inversion base is at the tropopause. The relationships of turbulence, flow speed, and mass emission load are comparable in the two cases. As in water pollution control, dispersal of the sources to prevent excessive concentrations before dilution can occur would be a useful control measure. Likewise, timed emissions of pollutants can be utilized to keep concentrations lower. Periods of low water flow produce higher concentrations of contaminants, and the same is true of low wind speeds in air pollution. The air pollution problem can be considered to consist of four steps in sequence: production, emission, transfer, and reception (Frenkiel, 1956). During each phase, attempts can be made to reduce the danger of contamination and in most cases scientific and technological methods already exist. In some cases it is possible, by changing production methods, to reduce the discharge to the air. In others the production methods may be changed in order to attach air pollution control equipment, thus reducing the emission. But in the general case air pollution control equipment is considered to reduce the emission to the atmosphere without changing the production process.

Production classification

Aerial contamination may be grouped in several ways: by type of activity, by type of process, by type of contaminant. The most general classification is by type of activity, since one industry may use several processes and evolve many different contaminants. The general form as in the US Standard Industrial Classification used by the Census Bureau is a useful grouping. Thus the total contaminant load per industry, e.g., petroleum refining, asphalt paving, rock crushing, is available for comparison. General pollution sources other than industrial are those resulting from traffic and incineration of rubbish and waste materials.

A more specific classification of pollution sources is by process. Here, cross-industry tabulations of processes allow one to assess the relative importance of industrial procedures and efficiency of available control devices. Assessment of contributions to air pollution by combustion, evaporation, grinding, crushing, etc., can be made. Of greatest importance is the classification by type of contaminant. In this instance, emission rates in terms of average and maximum values are needed in order to estimate pollution levels after dilution, downstream at the point of damage or perception. Involved in these considerations are the state of the pollutant, whether gaseous or particulate, its relative toxicological or nuisance property, its additive or synergistic effects, and exposure of the recipient. More recently attention has also been given to the chemical properties of certain pollutants, as various aerial reactions have been shown to occur, for example, in the atmosphere of the Los Angeles area.

Assumed in the production classification is the fact that a source, even though controlled, may still emit some pollutants to the atmosphere. Actually, if all source controls could be absolute, there would be no need for further discussion, as the problem of air pollution would not be a factor in site selection or zoning.

Transfer and perception

Once the pollution has been emitted from the source, the mechanism of transfer must be considered. The meteorological information of the general region yields gross information as to the extent to which air pollution sources need to be controlled. Mesometeorological data bearing on the particular site under consideration determine the degree and/or nature of control for that site.

It is conceivable that at times the importance of the industry or process which produces the air pollution may cause abandonment of certain practices or land uses in the immediate area. For example, consider the change in the agricultural pattern in the immediate Los Angeles vicinity, where smog-sensitive leafy vegetables are no longer grown on a large scale. The " green belt area " principle advocated by planning authorities in Great Britain separates industry and its emissions from the human perceptors (Howard, 1946), and in the USSR a similar suggestion has been made to remove the inhabitants from an area two kilometres in radius surrounding refineries and other specified industrial activities, thus creating a buffer zone (Goldberg, 1948).

In the discussion to follow, site selection and zoning will be considered in close relationship. Zoning is the application by regulation of the planning programme which encompasses all the sites in a particular area or region. Planning and zoning are therefore matters of official concern. By contrast, site selection is primarily the concern of the industry seeking to locate a plant within a given area.

Site Selection as an Air Pollution Control Measure

Site selection is the process of locating a single industrial plant and is alternatively referred to as plant location. From the standpoint of air pollution control, site selection results in the creation of a single source of pollution as compared with the more complex problem of area or multi-source emission (Larson, 1956).

In the USA, site selection is mainly the concern of the individual industrial plant and is influenced primarily by economic factors. To establish intelligent and reasonable control regulations, governmental agencies must understand the factors involved in selection. As in most human endeavours, there is a wide discrepancy between actual practice and accepted theories of site selection.

Stated otherwise, the theory of site selection is still in the developmental stage and consists of hypotheses not yet verified or substantiated.

The best summary of current American site selection theory is by Woodbury (1953) in his chapter on industrial location and urban redevelopment. Outlined therein is Hoover's basic framework of industrial location. The assumption is made that site selection is concerned with finding the best location, keeping expenses low, and income at the highest level possible. It recognizes two general types of cost, processing and transfer. Processing costs include on-site fabrication expenses, while the relatively more important transfer costs include expenses of bringing in raw materials and delivery of finished products. Four major types of industry are identified: material-, labour-, and market-oriented, and " foot-loose " industries. The advantages of the large urban area such as labour supply facilities, services, marketing and related industries are noted. The history of industrial site selection has developed in two phases. The first was the formative process where it depended heavily on the existing central city, and the second is the movement to outlying fringe areas of branch plants of well-established firms or a single-nature firm with expansion requirements. Site selection apparently proceeds in two steps. The first is the selection of a general region, and the second is the choosing of a specific site within the region. Surveys show that the region is selected on a more objective or rational basis than in the latter phase of picking a specific site (Dickson, 1948, Garrabrant, 1953; Los Angeles County Chamber of Commerce, 1954). In relation to air pollution control the considerations should be in reverse order, as wide variations of climatic features can occur within an economically advantageous region.

Normal factors of significance in selection of a general region are: markets; labour; material; transportation costs; utilities; climate and living conditions. Specific local site selection factors are: availability of usable land; building availability or costs; proximity to labour, housing, transportation, utilities, and related industries; plant protection, facilities of police and fire service; amenities such as housing, recreational and cultural facilities for employees; and restrictive zoning, water, and air-pollution regulations (Atkins, 1952; Breese, 1954; Newhoff, 1952; US Department of Commerce, Area Development Division, n.d.).

Air pollution factors

Factors related to air pollution have acquired considerable importance in recent years. Private enterprise has been forced to a broader view in site selection by the realization, whether influenced by law or by other pressures, that such selection should not have a degrading effect upon the community. To avoid costly control measures, to improve public relations, and to prevent litigation, Larson (1956) recommends that site selection take into consideration the nature of the air contaminants, the efficiency of available control devices, pertinent meteorological factors, and the potential effects on the surrounding areas. For instance, in the location of sites for atomic reactors, weather is a most important factor (White & Pack, 1956). Site location in these cases has had to consider not only the effluents from the atomic disintegration process, but also the possibility of accident resulting from a runaway reactor (US Weather Bureau, 1955). Continued experience and improved techniques will lessen the likelihood of accidents and will allow the reactors to be located closer to urban developments. The requirement of many industrial processes for supplies of clean air introduces another aspect of air pollution into the process of site selection. For example, clean air is necessary for cooling the reactors of atomic energy plants, since if polluted air were used, the impurities present would become radioactive and their escape into the atmosphere would create a hazard. Many micro-assembly plants need extremely clean air in order to maintain product quality. One such type of industry is the manufacture of transistors. Also, manufacturers of medical items such as antibiotics, vaccines, and sera must maintain practically sterile standards of air purity. In these cases, location of industries in areas of heavy air pollution will add materially to the cost of cleansing the air.

The prime factors to be considered in efforts to minimize air pollution problems by site selection are the climate and meteorology of the locations under consideration. The dispersive ability of the air at each possible site has to be determined on the basis of average values for wind movement and inversion conditions. Again the similarity to water pollution must be pointed out. In order to disperse effectively the pollution load, the air has to dilute the pollution load down to acceptable levels of contamination. Acceptable levels will have to be determined for each community or region. Hewson (1957) describes the two-step process " general region and specific location" for analysing the meteorological environment necessary for favourable atmospheric diffusion.

The general regional characteristics may be extracted from the long-term climatic record at a representative weather station. The local mesometeorological features may be inferred from analysis of the topography; soil; degree, type, and location of vegetation cover; location of built-up areas, etc. If such local data are not definitive, then a programme should be initiated to pinpoint local factors influencing air pollution dispersal. Moreover, even if a site is already chosen, ambient contaminant levels should be measured for at least a year to determine variations of pollution and provide a basis for estimating future contaminant levels.

Individual sources may be located in close proximity to each other without their emissions becoming additive, provided the local air flow passes only infrequently from one to the other. The airflow, or trajectory, is influenced greatly by the topography in the neighbourhood of the site under investigation. Terrain features such as location in a valley or near other orographic features which channel the airflow are readily recognized as predominant features and thus are determining factors in the trajectory configuration. Also of importance is the location of sites on moderate land slopes in relation to pollution receivers, as drainage winds may move emitted contaminants over very sensitive receptors. Certain insecticides are known to sensitize vegetation to the effects of smog of the type found in the Los Angeles area. Emissions from an insecticide manufacturer may have an inhibiting effect on agriculture when field crops dependent upon bees for pollination are located downwind. Similarly, 2-4 D manufacturers have experienced difficulty when broad-leafed vegetation exists downwind from the site or from the point of spray application.

In considering each possible site for the location of a plant, the industry has to balance the factors relating to availability and cost of control of air pollutants against site advantages. Local regulations may require controls so efficient that the expense of compliance may out-balance favourable cost factors due to market proximity, labour availability, etc. A government jurisdiction imposing these pollution restrictions must also be aware of controls available and not impose conditions impossible of accomplishment. The control of automobile exhaust would certainly lower the atmospheric pollution load in communities such as Los Angeles where this mode of transportation is predominant. However, at present there is no proven method for source control of automobiles short of their abolishment. Thus the curtailing of automotive use, while a social benefit from the standpoint of air pollution control, would immobilize the community and be an achievement gained at the expense of the entire community. Power plants are another example of the balance that has to be achieved between the power requirement for a community and the pollution resulting from its production. Both of these illustrations show the need for intelligent application of air pollution controls *per se*, or in the form of zoning requirements, in order that the growth of a community will not be hampered. The regulatory authority should establish:

(*a*) that it is necessary to reduce the pollution load,
(*b*) that control equipment exists, or the control principle is known,
(*c*) that the requirement for its use will not be discriminatory, and
(*d*) that it is feasible in terms of total economics versus achievement.

Basis for Zoning as an Air Pollution Control Measure

As pointed out, air pollution progresses in four steps: production, emission, transfer, and perception. Land use planning in the form of zoning may be applied to each phase. The location of the sources of production, the amount to be emitted, and the location of land uses susceptible to air pollution damage lend themselves directly to the principles of planning and zoning. Zoning as a control measure is based mainly upon a knowledge of the mechanics of the atmosphere, which is the transfer medium. To develop a comprehensive regional plan of land utilization a complete inventory of the demographic, topographic, and climatic features of the area concerned is necessary. These three items are listed in decreasing order of man's influence upon them. The climatic features are not controllable at all, but their relationships to air pollution must be taken into account. Minor changes in topographic features are now within the capabilities of man, but changes on a regional basis to cause alterations in climatic conditions are still not practicable. Demographic factors are wholly dependent upon the utilization or occupancy of the area under study.

Climatic features

The weather factors most directly determining the capability of the atmosphere of a particular region to disperse air pollution are air stability and wind movement. Wind data are observed continuously, while the air stability is measured periodically. During interim periods of stability determinations the change in stability is followed by observing the surface temperature. Wind data are usually readily at hand, while stability data, being more costly to obtain, are usually lacking in critical area.

The weather element which reflects the intensity of air pollution is visibility. Weather stations that measure surface wind velocity normally also report the visibility and the condition which limits the visual range at low values. Evaluation of trends of daily average visibility or numbers of days per month of low visibility for extended periods often reflect the urbanization of the surrounding area. Usually these data are only collected after the onset of an air pollution problem, and comparison to pre-pollution periods is impossible. If sufficient visibility reporting stations are established within the region concerned, it may be possible to evaluate the influence of single industrial plants or types of plant. Increases in visibility at weekends are indicative that plants such as refineries, steel mills and the like, which maintain operations seven days a week, have little effect on reducing visibility on the week days.

The atmosphere exhibits self-scavenging ability with respect to air pollutants, similar in principle to pollution elimination in flowing water. Oxidative processes tend to reduce organic pollution to carbon dioxide and water vapour. Aerosols conglomerate and settle out or are washed out by rainfall. Pertinent climatological considerations range all the way from world-wide phenomena to highly specific information related to specific sites. Climatological analogies may be recognized in widely scattered regions. For example, the situation confronting Los Angeles and vicinity, in which general circulation patterns produce prolonged subsidence inversions, occurs frequently in at least four other regions of the world; industrialization of these areas should be discouraged if alternate localities are available. At present these other areas—the north-west coast of Africa, western coast of South Africa, south-west coast of South America, and east coast of Australia—have relatively little industry.

Geographical and topographic features

A noticeable characteristic of modern industrial plants is the trend to one-storey construction, which demands nearly level terrain for economic building and ease of transportation access. It has been found that land slopes of less than 10% are advantageous for modern industrial plants. A land-slope study should be accomplished for the region concerned and account taken of existing land uses which cannot be supplanted by industry. In the past it has been usual for zoning officials to designate residential and commercial areas and then relegate remaining areas to an industrial classification (National Industrial Zoning Committee, 1951). Thus, in spite of an apparent abundance of industrially zoned land, owing to the zoning of submarginal land by default, there may be an actual shortage of usable industrial sites (Muncy, 1954). This lack of adequate industrially zoned land is shown by constant modification or amendment of zoning regulations to increase the industrial areas in spite of existing vacant industrially zoned properties. Industries extracting natural resources such as rock, gravel, sand, oil, coal, etc. must be located where the resources are placed by nature. Land zoning in the vicinity of these resources must take into account the associated aerial contamination which will result, and, as the source cannot be moved, pollution receptors should be so located that they will not be adversely affected.

Owing to large-scale pressure and temperature gradients larger mountain chains do not restrict the flow of pollution, but they do channel the direction of the flow into and through valley passes. Smaller areas of concern are individual valleys where localized wind movements may occur, and prolonged periods of nearly calm conditions are frequent. Industrialization of these areas must be minimized.

Economic and cultural features

In order to predict future circumstance it is necessary to extrapolate from current and past knowledge. The existing industrial pattern must be inventoried and classified before alternative future programmes can be recommended. Industrial data are normally presented only in summary form without regard to location within defined areas. Usually the boundaries of these areas are related to jurisdictional or governmental factors and change from time to time. The survey method adopted should provide for comparability of data. A one-mile grid system has been adopted by the Los Angeles County Air Pollution Control District as a means of locating factories independently of changing local governmental boundaries. The grid is also used for trajectory analyses, isodemic correlations, traffic flow and other related demographic features. Since few individual manufacturing concerns can afford to conduct surveys on a regional basis, such studies will need to be underwritten by governmental agencies or by trade associations.

The location of existing industrially zoned land may not be sufficient for the future. Current residential areas may deteriorate to blighted areas, and may be redeveloped as industrial. To find the areas which may be expected in the future to house industry, the " sieve " procedure developed in Great Britain in 1938 may be used to eliminate that land which is not suitable for industrial use because of such factors as excessive slope, lack of access, scarcity of water, or other limiting factors of the areas (National Resources Planning Board, 1942). Only the vacant land in this remaining area can be considered as potential industrial land. Some previously occupied areas have been redeveloped for industrial use in the USA under the provisions of a Federal Act, but this has occurred only in limited areas (Garrabrant, 1955). This method of eliminating blighted areas was undertaken because zoning *per se* was relatively ineffective. There are several demographic features which should be considered in forecasting the industrial growth of the community. Perhaps the most readily at hand is the present location of the population, which can be shown in terms of an isodemic map of the area under study. By constructing similar maps for previous census enumerations, a graphic presentation of the past growth is developed. Extending this to the future will yield isodemic maps which can be related on a per capita basis to area requirements for industrial acreage and may also form a basis for deriving traffic loads between expected resident population centres and forecast employment centres. At the present time the automobile exhaust, although recognized as a major contributor to air pollution, is without a control mechanism. Until the control of this pollution source is resolved, traffic itself must be considered a source as well as a result in applying zoning principles to the air pollution problem.

Applications of Zoning to Control of Air Pollution

Whenever the pollution level becomes critical owing to ineffective voluntary control efforts, the public demands, and usually receives, compulsory regulation with more restrictive or arbitrary controls than those which existed previously. Under such conditions zoning may take the sterner course and simply prohibit offending industries from the entire area or from portions of selected areas. Such drastic measures may not be the ideal solution for large metropolitan areas but may be the answer for some satellite cities. A less restrictive policy may be one providing open areas, preferably government owned, for multipurpose use, such as watershed, flood control, airport approach, recreation, to allow greater dilution of fall-out of contaminants before they reach the receptor areas. If the contaminants are not harmful to vegetation, then the open areas may take the form of the green-belt development advocated by city planners.

Source density

Density of industrial source regulations may be applied directly as source or sources per unit area, to keep emission concentrations within acceptable mass concentrations (Larson, 1956). Indirect density regulations are available through minimum or maximum yard, height, size of site, workers per acre land coverage, and building bulk requirements. Moreover, through public policy, additional means may be used to further the decentralization of industries through assessment, rate setting, and circulation policies. Conversely, the low density provisions may be applied to restrict receptors of smog damage such as residential areas in the path of high smog concentrations.

An alternative to dilution by dispersion through mechanical and thermal turbulence is the dispersion of the source itself or decentralization of industry. Further indirect air pollution benefits might be derived from the possible reduction in automobile exhaust if industry were decentralized to the residential suburbs of the metropolitan area (Warren & Davidge, 1930). Since Los Angeles residents travel predominantly by automobile, it is understandable that if the residents were to work in nearby factories, air pollution from this source would be reduced.

While a legal authority to zone with respect to air pollution control may exist, the application of such power must not be arbitrary, capricious, or unreasonable. A reasonable basis for the zoning must exist and may take the form of a mesometeorological study showing the inter-relationship of areas within a region due to wind trajectory frequencies and degree of pollution. This system of allowing or prohibiting industries in specific areas will only work if the mesometeorological study shows channelling effects in the wind movement, especially during periods conducive to air pollution accumulations. When the situation of haphazard wind movement arises, then the system of intra-regional placement will not be effective and source location density will have to be restricted to obtain acceptable pollution levels.

The continuing influx of population into urban centres is giving rise to super-cities, and the proper location of industries will be hampered owing to the division of control between many governmental units. The jurisdictional area for control must embrace all the urban area subject to the climatic features which induce the air pollution problem. Within a climatic region, zoning based on the local weather factors would allow for the " districting " of sources in conformity with an over-all plan. This placement policy would supplement the present system of control at the source.

Timed emissions

The principle involved in timed emissions is the imposition of two sets of performance standards in a locality where air pollution problems exist interspersed with periods of low and high dispersion capability (Hewson, 1957). The higher performance standards would be required at the onset of weather conditions conducive to air pollution. The Trail, British Columbia, incident resulted in an example of source control during periods when pollution accumulates, with no source controls when conditions of no inversion and relatively high winds do occur. In Los Angeles County this principle has been applied to the problem of restricting rubbish burning in the southern portion of the County, dependent on whether the dispersive volume defined by certain meteorological parameters is below critical values. The Los Angeles County Air Pollution Control District also has a regulation relating to emergencies when toxic contaminants in the atmosphere exceed specified levels. In this latter case the control becomes increasingly strict until the worst conditions occur and source controls require complete shutdown of all industrial pollution sources.

Specification of raw materials and processes

In certain critical areas another means of control would be through the regulation of raw materials and processes (American Society of Planning Officials, 1955b; O'Harrow, n.d.). For example, the type of fuel to be burned could be limited to natural gas, and residual fuel oil could be prohibited. Another example might be that no organic solvents would be permitted for cleaning and painting processes in specified areas. Smokeless zones have been created in selected industrial areas in Great Britain by limiting industries and residences in these zones to the use of certain specific fuels (Wilmott, 1952). With technological advances, the substitution of materials and chemicals may be more feasible in the future. In the Los Angeles area, the inefficient single-chamber incinerator has been outlawed for industrial and commercial uses and will soon be prohibited entirely, allowing only the more efficient multiple-chamber incinerators.

Specification of fuel composition can be accomplished by zoning ordinances, and this principle has been applied in St Louis and Pittsburgh and other areas where the use of high volatile fuel is prohibited. The oil industry is continuously improving the yield of light fuels per barrel of crude, with the result that impurities, such as sulfur, are concentrated to a smaller residue. Although this residual tarry oil is a relatively inexpensive fuel, its high sulfur content gives rise to visible plumes which may restrict visibilities over large areas. This type of operation is quite common when electric power plants are constructed near refineries. Zoning may apply in this case by restricting the sulfur content of the fuel to be used to a level where the emissions are no longer bothersome. An alternative source of energy in areas where power is necessary and pollution problems are critical is afforded by recourse to atomic power. In the atomic power plant there is no large volume of combustion exhaust gases to scrub for dilute concentrations of contaminants and control measures are feasible.

REFERENCES

American Society of Planning Officials (1955a) *Air pollution—a growing urban problem*, Chicago, p. 31

American Society of Planning Officials (1955b) *Industrial zoning standards*, Chicago, p. 1

Association for Planning and Regional Reconstruction (1950) *Town and country planning textbook*, London, p. 66

Atkins, R. M. (1952) *Harvard Business Rev.*, **30,** 113

Breese, G. (1954) *Industrial site selection*, Princeton, p. 2

California State (1955) *Laws relating to conservation, planning, and zoning*, Sacramento, p.18

California University, Bureau of Public Administration (1957) *Adapting government to metropolitan needs*, Sacramento, p. 43

Chute, C. F. (1955) *A new approach to the problem of government in metropolitan areas.* In: *Talk presented before the national conference on government*, Seattle, p. 11

Dickson, P. W. (1948) *Decentralization in industry*, New York, p. 9

Frenkiel, F. N. (1956) *Sci. Monthly (Wash.)*, **82,** 194

Gallion, A. B. & Eisner, S. (1950) *The urban pattern*, London, New York, Toronto, p. 227

Garrabrant, R. B. (1953) *The community and industrial development*, Washington, p. 9

Garrabrant, R. B. (1955) *Redevelopment for industrial use*, Washington, p. 23

Goldberg, M. S. (1948) *[The sanitary protection of air]*, Moscow, p. 119

Hewson, E. W. (1957) *Purdue Univ. Engng Bull.*, **41,** 213

Hoover, E. M. (1948) *The location of economic activity*, New York, Toronto, London, p. 265

Horack, F. E., jr & Nolan, V., jr (1955) *Land use controls*, St Paul, p. 64

Howard, E. (1946) *Garden cities of tomorrow*, p. 2 (F. J. Osborn), p. 32 (L. Mumford), and p. 55

Larson, G. P. (1956) *J. Air Poll. Control Ass.*, **5**, 211

Lieberman, J. A. (1957) *J. publ. Hlth*, **47**, 345

Los Angeles County Chamber of Commerce (1954) *Survey of 118 plant locations in Los Angeles County*, Los Angeles pp. 5 and 10

MacDonald, A. (1956) *Industr. Wastes*, **1**, 91

Muncy, D. (1954) *Harvard Business Rev.*, **32**, 54

National conference on metropolitan problems, Proceedings of (1956) New York, pp. 47 and 62

National Industrial Zoning Committee (1951) *Principles of industrial zoning*, Columbus, p. 2

National Resources Planning Board (1942) *Industrial location and national resources*, Washington, p. 348

Newhoff, M. C. (1952) *Trends in industrial location*, New York, p. 6

New York State Department of Commerce (1953) *Zoning in New York State*, New York, p. 9

O'Harrow, D. (n.d.) *Performance standards in industrial zoning*, Columbus, p. 4 (National Industrial Zoning Committee)

United States Department of Commerce, Area Development Division (n.d.) *Basic industrial location factors*, Washington, p. 2

United States Weather Bureau (1955) *Meteorology and atomic energy*, Washington, p. 1

Warren, H & Davidge, W. R. (1930) *Decentralisation of population and industry*, London, p. 146

White, F. D. & Pack, D. H. (1956) *J. Air Poll. Control Ass.*, **6**, 151

Wilmott, T. E. (1952) *Smokeless Air*, **22**, 120

Woodbury, C. (1953) *The future of cities and urban redevelopment*, Chicago, p. 103

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PREVENTION AND CONTROL OF AIR POLLUTION BY PROCESS CHANGES OR EQUIPMENT

Introduction

Atmospheric waste products may be classified in two broad categories: aerosols and gases. Aerosols are liquid or solid particulates in suspension in a gaseous medium, usually air. In the field of air pollution these particulates are more specifically classified in terms of their method of generation, each class being further characterized by a particle-size range. Dusts are solid particles formed by some disintegration process such as crushing, grinding or demolition. Normally dust particles range above 2 μ in diameter but have been found to be as small as 0.1 μ. Fumes are solids generated by the condensation of vapours and may result from sublimation, distillation, or foundry processes, or from chemical reactions. The normal particle size of a fume is less than 1 μ. Fumes are often metals or metal oxides and their composition may differ materially from the substance from which they originated. Mists are liquid droplets smaller than 10 μ in diameter and are generated by condensation. Sprays are larger liquid droplets which are created by some mechanical disintegration process. In the collection of liquid or solid aerosols, chemical identity is not normally a factor in the choice of a control technique; however, in the control of gaseous emissions, chemical properties are often paramount in developing the technique employed.

The degree of source control necessary to overcome a specific area's air pollution problem is a function of the mass rate of emission of contaminants to the atmosphere within that area, the particular source concentrations present, the meteorological characteristics of the area, and the area's air pollution susceptibility.

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Mass rates of emission and source concentration may be estimated or actual source samples may be taken. In order that correlations of mass rate of emission and source concentrations may be more applicable, source discharges should be expressed as a function of some unit of production, e.g., weight loss per unit of throughput. Air pollution sources may be classified as domestic and industrial combustion processes, automotive engines, petroleum processing operations, chemical manufacturing processes, pyro- and electrometallurgical processes, mineral processing and food and feed manufacturing operations. Table 1 gives the type of contaminant generally discharged to the atmosphere from typical sources in each of these broad classifications. It is important to recognize that while practically all such operations lead to the discharge of air pollutants, quantities within a single industrial category will vary as a result of production methods in use, raw materials employed, and air pollution control measures utilized.

TABLE 1

SOURCES OF AIR POLLUTION

Class	Aerosols	Gases and vapours	Typical loss rates
Combustion processes	Dust, fume	NO_2, SO_2, CO, organics, acids	0.05 % - 1.5 % by weight of fuel
Automotive engines	Fume	NO_2, CO, acids, organics	4 %-7 % by weight of fuel (hydrocarbons)
Petroleum operations	Dust, mist	SO_2, H_2S, NH_3, CO, hydrocarbons, mercaptans	0.25 % - 1.5 % by weight of material processed
Chemical processes	Dust, mist, fume, spray	Process-dependent (SO_2, CO, NH_3, acids, organics, solvents, odours, sulfides)	0.5 % - 2 % by weight of material processed
Pyro- and electro-metallurgical processes	Dust, fume	SO_2, CO, fluorides, organics	0.5 % - 2 % by weight of material processed
Mineral processing	Dust, fume	Process-dependent (SO_2, CO, fluorides, organics)	1 % - 3 % by weight of material processed
Food and feed operations	Dust, mist	Odorous materials	0.25 % - 1 % by weight of material processed

The second factor—meteorological characteristics of the area—which must be evaluated in determining the degree of industrial source contaminant control necessary affects the rate of accumulation and dispersion of air contaminants for that specific area. The dispersion of area concentrations of atmospheric pollutants by air motion is a continuous process but the rate at which the contaminants are dispersed is variable and depends on the horizontal and vertical movements of the atmosphere.

Finally, consideration must be given to the area's air pollution susceptibility; that is, certain areas have a greater potential for air pollution than others. In this sense air pollution may be considered as an atmospheric condition defined in terms of its identifiable manifestations, i.e., irritation of the eyes and mucous membranes, reduced visibility, and damage to plants, animals, and property.

Basically, four procedures are available for the control of effluent discharges to the atmosphere and thereby of their detrimental effects. These are: (*a*) reduction of contaminant discharge at the source by application of control equipment; (*b*) reduction at the source through raw material changes, operational changes or practices, or modification or replacement of process equipment; (*c*) dilution of the source discharge by the use of tall stacks; and (*d*) dispersion of source locations through allocation of land usage.

It is the purpose of this discussion to consider only the first two of these techniques.

The Application of Control Equipment for the Prevention of Air Pollution

The objectives sought for in the application of air pollution control equipment are the prevention of nuisance or physical damage to property, the elimination of health or safety hazards to plant personnel and to the general populace, the minimization of economic losses through the reduction of plant maintenance, the recovery of valuable waste products, and the improvement of product quality.

Performance of air pollution control equipment is assessed in terms of collection efficiencies. For particulate collection, efficiency may be defined as percentage removal by weight, percentage removal by weight per particle size range, or percentage removal by count. Evaluation of equipment effectiveness may also be accomplished by the measurement of the relative opacity of the discharge or simply in terms of the amount of contaminant released. Gaseous control effectiveness is normally measured in terms of discharge concentrations of the contaminating material.

In discussing the equipment used to control industrial air pollution at the source, it is convenient to divide it into two basic types—that for the control of solid and liquid aerosol emissions and that for the control of gaseous emissions.

Control of aerosol emissions

The general principles of dispersoid collection are applicable for both solid and liquid aerosols. While it is not the intent of this paper to review the fundamental theory behind these principles, sufficient theory will be presented to illustrate the removal mechanisms employed in each type of particulate collection equipment.

It is not uncommon for extremely wide ranges of particle size to be encountered in air pollution control work; also, concentrations of aerosols in industrial applications may range from a fraction of a grain per cubic foot[1] to about 20 grains per cubic foot and occasionally loadings as high as several hundred grains per cubic foot are reached. It is evident, therefore, that with such extreme variations of aerosol size and concentration, many types of collection equipment will be required to solve the industrial effluent control problem.

When considering the removal of dispersoids from a carrier gas stream, the following factors affecting the choice of a particular piece of control equipment are significant:

Particulate characteristics, including particle size spectrum, effective particle shape, particle density, and physico-chemical properties such as stickiness, hygroscopic properties, agglomeration tendencies, corrosiveness, flowability, electrical conductivity, flammability, toxicity, etc.

Carrier gas characteristics, including temperature, pressure, and physical properties such as humidity, dew points of condensable components, density, viscosity, electrical conductivity, etc., and chemical properties such as corrosiveness, flammability, toxicity, etc.

Process factors, including constancy or variability of gas flow, volumetric gas rate, particulate concentration, collection efficiency requirements, allowable pressure drop, and product quality requirements.

Operational factors, including maintenance, continuity of operation, safety and health protection, and ultimate use of collected materials.

Constructional factors, including structural limitations such as floor space and headroom, and material limitations such as pressure, temperature and corrosion service requirements.

Gravity settling chambers

The gravity settling chamber is the simplest type of equipment employed for the collection of solid and liquid particulate discharges. It consists of a chamber in which the carrier gas velocity is reduced so as to allow the dispersoid to settle out of the moving stream under the action of gravity. The efficiency of collection for particles having a terminal settling velocity, u_t, is expressed by the general equation:

$$E_g = \frac{u_t A_h}{Q} \qquad \text{for } E_g \leq 1.0 \text{ (consistent units)} \qquad (1)$$

where, E_g = weight collection efficiency of gravity settling chamber for particles of terminal settling velocity, u_t, dimensionless

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[1] 1 grain/cu.ft = 2.288 g/m³.

u_t = particle terminal settling velocity, cm/sec. or ft/sec.
A_h = projected horizontal area of chamber, cm^2 or sq.ft
Q = volumetric flow rate of carrier gas, cc/sec. or cu.ft/sec.

From this equation it can be seen that, for a given volumetric gas flow rate, the collection efficiency for any particle having a settling velocity of u_t depends on the total plan cross-section of the chamber and is independent of the height of the chamber. The minimum height of a gravity settling chamber, however, is established by the requirement that gas velocity through the chamber be sufficiently low as to prevent re-entrainment of the separated dispersoid. Normally this velocity should not exceed 10 feet[1] per second.

It is essential in this type of collector that the lateral velocity profile be as uniform as possible and that a low degree of turbulence relative to the settling velocity exist. Deviation from these requirements will result in reduced efficiency of collection. These criteria are normally achieved by employing gradual inlet transitions, guide vanes, or gas flow distribution plates.

Arrangements of vertical baffles have been used in some instances on the theory that increased efficiency will result from the superimposition of inertial forces on the particles. However, inertial effects from such an arrangement are so small that they are exceeded by the effects of increased velocity through the collector and may even result in decreased efficiencies.

From a practical standpoint, industrial application of this equipment is limited. Practically, gravity separators are not employed for the removal of dispersoids below 40 μ in diameter because of the excessive size of the equipment which would result for collection below this size limit. Even for the particle-size range above 40 μ, space requirements for this type of equipment are large.

A modification of the simple settling chamber employs a number of horizontal plates or trays within the chamber and this arrangement can lead to marked improvements in performance. As seen from equation (1), collection efficiency depends directly upon projected horizontal area of the collector. Hence, the increase in efficiency obtained by the insertion of horizontal shelves is directly proportional to the number of shelves. Even with such equipment, however, the minimum particle size which can be collected in practice is about 10 μ. Use of this type of modified settling chamber is limited by difficulties in cleaning closely spaced trays and by their tendency to warp during high temperature operation. Also, this type of collector is not practicable for handling dispersoid concentrations exceeding one grain per cubic foot.

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[1] 1 ft = 0.3 m.

Properly employed and designed gravity separators possess many inherent advantages: (*a*) low initial cost; (*b*) simple construction; (*c*) low pressure drop; (*d*) low maintenance requirements ; (*e*) dry and continuous disposal of solid particulates; and (*f*) temperature and pressure limitations imposed only by materials of construction used. These advantages, however, are offset to a degree by several major disadvantages: (*a*) large space requirements; (*b*) decreasing collection efficiencies with decreasing particle size (very low efficiencies for particles below 10-40 μ in diameter); and (*c*) decreasing collection efficiencies with decreasing inlet dispersoid concentration.

Because of simplicity of construction and low maintenance costs, gravity settling chambers have found quite widespread application as pre-cleaners for higher efficiency collectors. This reduces the inlet dust loadings to the second-stage cleaner and can remove large, highly abrasive materials, thus reducing maintenance costs of high efficiency equipment which is more subject to abrasive deterioration. Settling chambers are used widely for the removal of large solid particulates from natural draft furnaces, kilns, etc.

Inertial separators

This classification of control equipment includes all dry-type collectors which utilize the relatively greater inertia of the dispersoid to effect particulate-gas separation. Two types of equipment utilize this fundamental principle: cyclonic separators, which produce continuous centrifugal force as a means of exerting the greater inertial effects of the dispersoid, and simple inertial or impaction separators, which employ incremental changes of direction of the carrier gas stream to exert the greater inertial effects of the dispersoid. Because of the importance of cyclonic separators in the air pollution control field, they will be discussed in a subsequent section.

An obstructing body present in a moving dispersoid-laden gas causes the gas stream to be deflected around the body. The dispersoid, however, as a result of its greater inertia, will tend to cross the fluid streamlines and to impinge on the surface of the obstructing body. Impingement separation may be analysed theoretically in terms of so-called target efficiencies. Target efficiency is defined as the fraction of particles, in the fluid volume swept by an obstructing body, which will impinge on that body. Theoretical expressions for target efficiencies for simple geometries may be derived from classical hydrodynamics, but for more complex situations experimental determinations are required. It has been shown (Albrecht, 1931; Langmuir & Blodgett, 1946; Sell, 1931) that in the Stokes Law region, for any geometry, target efficiency should be some function of the dimensionless group,

$$[D_p{}^2 V_o (\rho_p - \rho)] / [18 D_B \mu]$$

where, D_p = particle diameter, cm or ft
V_o = velocity of fluid relative to obstructing body, cm/sec. or ft/sec.
ρ_p = absolute density of particle, g/cc or lb/cu.ft
ρ = density of fluid, g/cc or lb/cu.ft
D_B = characteristic dimension of obstructing body, cm or ft
μ = viscosity of fluid, poise or lb mass/ft sec.

For impingement of particles on an infinite cylinder or on a sphere, the following relationships hold theoretically:

$$E_t \text{ (cylinders)} = \frac{D^2_p\, V_o\, (\rho_p - \rho)}{18\, D_B\, \mu} \qquad (\text{for } E_t < 1.0) \quad (2)$$

$$E_t \text{ (sphere)} = \left[\frac{D^2_p\, V_o\, (\rho_p - \rho)}{18\, D_B\, \mu}\right]^2 \qquad (\text{for } E_t \leq 1.0) \quad (3)$$

where, E_t = target efficiency, dimensionless.

It may be said, therefore, that collection efficiency of an impingement separator increases with increasing particle size, with relative gas velocity, and with particle density; and decreases as the size of the individual impingement surfaces increases and as viscosity of the carrier gas increases.

These relationships have been derived for conditions of streamline flow; they should, however, be approximately correct for turbulent flow. Further, these relationships were developed for obstructing bodies in an infinite fluid and are applicable provided the adjacent collecting members are not so closely spaced as to cause appreciable distortion of the flow pattern; this condition is generally the case for commercial air filters and liquid scrubbers. Where the members are spaced such as to cause distortion of flow, the relationships for operating efficiencies will give conservative results (Lapple, 1950).

For the different types of equipment within this classification, the configuration of the obstructing bodies will vary from simple straight baffles to more complicated patterns which give maximum impaction efficiency with minimum pressure drop. The wide variation of impingement surface configuration employed in commercial units prohibits a detailed discussion of each. However, the more fundamental types, i.e., baffle type, orifice impaction type, high velocity gas reversal type, and louver type, will be discussed.

Perhaps the simplest type of impingement separator is the baffle chamber. Such a device forces the gas stream to follow a tortuous flow path, which is obtained by the insertion of staggered plates or shaped obstacles into the gas stream. In such devices the gas is forced into a series of sudden changes of direction with resulting impaction of particles on solid surfaces. This equipment is capable of removing particles larger than 20 μ in diameter with pressure drops varying from 0.5 to 1.5 inches[1] of water, depending upon the type of application and the configuration of the impingement elements. Frequently commercial units are equipped with some mechanism for cleaning the impaction surfaces. Rappers are sometimes provided to remove the collected materials; other units employ a continuous or intermittent flowing film of water over the collecting surfaces.

Because of the simplicity of construction, these units are capable of operating at elevated temperatures and pressures. However, care must be exercised that the materials to be separated are not tacky, since build-up on the impingement element can change the efficiency characteristics of the unit. Also, owing to the high impaction velocity in effect in this unit, abrasion is often critical. Baffle type units have found wide application for solids removal in power plants and rotary kilns and for the removal of acid mists.

The second type of inertial separator is the orifice impaction type composed of two groups of orifice plates in series. The first group consists of orifice plates followed by impact plates whose apertures are staggered from those in the first plate. Passage of the gas stream through this assembly promotes agglomeration of the conveyed particulates. The second group consists of several plates with aligned orifices and acts as the collector element for the agglomerated particles. Many modifications of this type are available on the market but all employ the general principle of orifice impaction for the agglomeration and collection of the dispersoid. High efficiencies have been obtained in this type unit for liquid aerosols larger than 2 μ in diameter. Normal velocities through the orifices are about 50-100 feet per second and seldom exceed 150 feet per second. The pressure drop through such units is approximately 2.0-2.5 orifice velocity heads.

The third class of inertial separators is the louver type (Fig. 1). This unit contains a series of louvers or impingement elements set at an angle to the air stream so as to cause a single rapid reversal of air flow direction and thereby cause the dispersoids to impinge on the louvers. The particles thus impinged rebound back into the moving air stream in the inlet chamber and are removed from the collector by a secondary air circuit. Efficiency of this type of unit is basically a function of the louver spacing, closer spacings producing higher efficiencies. Plugging due to build-up on the front and back sides of the louver or by mechanical obstructions will tend to reduce efficiency and volumetric capacity. For practical applications, efficiencies for this type of unit are similar to those for other dry-type inertial collectors. The principal advantages of this type of collection equipment are simplicity and low cost of construction as well as moderately low pressure drop for the degree of removal obtained. The disadvantages are a tendency for

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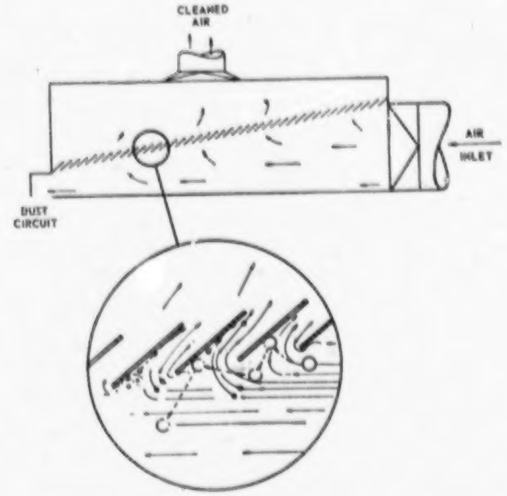
[1] 1 inch = 2.54 cm

FIG. 1
LOUVER TYPE COLLECTOR



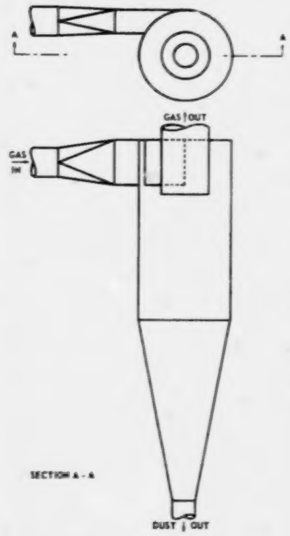
accumulation of particles on the louvers with a corresponding reduction in efficiency and excessive abrasion of the louver elements, as well as an inability to handle tacky materials. Temperature and pressure limitations are again imposed only by the materials of construction.

In contrast to the above types of separator, the high velocity gas reversal chamber employs inertial separation, without impaction, for the removal of particulate matter. Here the bulk gas stream is caused to change direction at high velocities, thereby projecting particulate matter into an appropriately provided dead air space, from which the dispersoid is removed by gravity. Efficiencies for this type of unit are low for particle sizes of less than 50 μ. This type of installation is sometimes used as a pre-cleaner for more efficient control mechanisms to reduce the load of large diameter particles to these units.

Cyclonic separators

The cyclone separator represents one of the least expensive and most important types of dispersoid collector. It commonly consists of a cylindrical or conical chamber with provisions for the entry of the dispersoid-laden gases tangentially at one or more points, and for discharge through a central cylindrical opening at the top (Fig. 2). The gas path generally follows a double vortex with the gas spiralling downward at the outside periphery of the body and upward through the outlet cylindrical section.

FIG. 2
LONG, SLENDER SINGLE CYCLONE



Owing to the rapid spiralling movement of the gas, the dispersoids are projected to the wall by centrifugal force and they then drop by gravity to the bottom of the body, where they are removed. During cyclonic separation carrier gas rotational velocity may exceed by several times the average inlet gas velocity.

On the basis of analyses analogous to those for gravity separation, in which gravitational acceleration is replaced by centrifugal acceleration, a number of equations have been developed for the minimum diameter of particle which can theoretically be completely separated from the gas stream (American Petroleum Institute, 1955). The following is a typical equation illustrating the parameters involved (Rosin, Rammler & Intelmann, 1932):

$$D_{p,\,min} = \sqrt{\frac{9\mu\ B}{\pi V\ N_t\ (\rho_p - \rho)}} \qquad (4)$$

where, $D_{p,\,min}$ = diameter of smallest particles completely collected, cm or ft
μ = viscosity of fluid, poise or lb mass/ft sec.
B = width of cyclone inlet, cm or ft
V = average inlet velocity, cm/sec. of ft/sec.
N_t = number of turns made by gas stream in cyclone, dimensionless
ρ_p = absolute density of particles g/cc or lb/cu.ft
ρ = density of fluid, g/cc or 1b/cu.ft

An extension of this analysis has been made leading to a method for the prediction of over-all cyclone efficiencies (Lapple, 1951). This simplified analysis assumes that the gas path within the cyclone is in the form of a rigid spiral and that the rotational velocity equals average cyclone inlet velocity. In practice the flow pattern is considerably more complex leading to deviations from the efficiencies theoretically predicted (Lapple, 1950). Other studies have indicated that a " double eddy " at the inlet to the cyclone is superimposed on the double spiral and may in some instances affect dispersoid removal (Magill, Holden & Ackley, 1956; Shepherd & Lapple, 1939; Van Tongeran, 1935; Wallman, 1938).

The design factor having the greatest effect on collection efficiency is the cyclone diameter. For a given pressure drop, the smaller the diameter of the unit the higher the collection efficiency obtained, since centrifugal acceleration increases with decreasing radius of rotation. Centrifugal forces employed in modern designs vary from 5 to 2500 times gravity depending on the diameter of the cyclone. Present trends have been toward smaller diameter cyclones; however, with such units, in order to maintain adequate volumetric throughput, multiple units in parallel have been employed. For a given design, the effect of increasing gas throughput and, hence, increasing the inlet velocity, is normally to increase efficiency, but at the expense of increased pressure drop. For practical applications cyclone design must therefore be limited by pressure drops which are feasible with commercially available fans. This limitation usually dictates that inlet velocities be in the range from 20 to 70 feet per second, but equipment is normally designed for a velocity of 50 feet per second. Normal present-day practice has established pressure drops for modern cyclones from 1 to 20 inlet velocity heads depending upon the geometric proportions of the equipment and upon the collection efficiencies required.

Further efficiency increases are thought to result from an increase in cyclone body length but there are no conclusive experimental data to verify this. It has also been proposed that a decrease in the diameter of the gas outlet produces a more significant effect on increase in efficiency (Linden, 1949). No general agreement has been reached as to the use of baffles, guide vanes, etc. for increasing cyclone efficiency. The predominant opinion is that, in general, they reduce efficiency (Alden, 1940; Lapple, 1950; Shepherd & Lapple, 1939, 1940).

Design results for cyclonic separators are no better than particle-size distribution data available to the designer, and less than rigorous attention to these data often results in unsatisfactory operation. Although there is considerable information in the literature on particle-size analysis, it has been noted that in practice data have sometimes been used which are statistically misleading and which have little physical significance when applied to cyclonic separator design. The measurement of particle size is not generally an absolute process and the method of measurement should, therefore, be based on the physical characteristics about which the information is desired. If, as in the case of cyclonic separator design, the size information desired is concerned with hydrodynamic and aerodynamic properties, the method should take into account particle mass, aerodynamic drag, etc.—i.e., terminal settling velocity. As a consequence, elutriation or sedimentation methods will give the more useful results for the design of cyclonic separators.

In practice cyclonic separators may be designed for the satisfactory collection of particles over wide ranges of size and concentration and over wide ranges of pressure and temperature. Cyclones have operated at temperatures as high as 1000° C and at pressures reaching 500 atmospheres. Satisfactory separation efficiencies, when employing the high efficiency, small diameter cyclones, are commonly obtained for dispersoids of 5-10 μ in diameter and greater. In applications where high dust concentrations, of the order of 100 grains per cubic foot and greater, are encountered, correspondingly high collection efficiencies for smaller particle sizes are obtained. In this regard it is significant that increased dust loadings will result in decreased pressure drop and increased collection efficiencies (Drijver, 1937; Shepherd & Lapple, 1939).

Cyclone designs are developed covering a wide range in size, geometry, and method of gas entry. Either cylindrical or conical cyclone bodies are employed with the diameter varying from a few inches, in the long slender units with high efficiencies in the sub-sieve particle-size range, to 15-20 feet in the short, large diameter units employed in the ultra-sieve particle-size range. Gas entries may be involute or tangential or may employ guide vanes to impart helical motion to the gas stream.

A variation of the simple cyclone design consists of units in which the centrifugal forces are developed by a mechanically driven rotating element. These units are so designed as to concentrate the dispersoid at the periphery of a scroll, housing the mechanical rotating element, where the dispersoid is removed through an annular slot or a skimmer. The chief advantage of the mechanical centrifugal separator lies in its compactness. Such units appear to have collection efficiencies of the same order of magnitude as those of the small diameter, high efficiency cyclonic separators. A limitation of this type of unit, however, is a tendency of certain types of solids to build up on the rotating element, causing plugging of the unit or rotor unbalance.

Cyclonic separators have found widespread acceptance for the control of gas-borne dispersoids in such industrial operations as aggregate processing, cement manufacture, feed and grain processing, food and beverage processing, mineral processing, paper and textile industries, and woodworking industries.

Universal acceptance of this type of collector has resulted from certain inherent advantages: (*a*) low initial cost; (*b*) relatively simple construction; (*c*) dry and continuous disposal of solid particulates; (*d*) low pressure drop; (*e*) relatively low maintenance requirements; and (*f*) excluding mechanical centrifugals, temperature and pressure limitations imposed only by materials of construction. The application of this type of separator is restricted, however, by three major disadvantages: (*a*) low collection efficiencies for particles below 5-10 μ in diameter; (*b*) decreasing collection efficiencies for decreasing dispersoid concentrations in the gas stream; and (*c*) equipment is subject to severe abrasive deterioration.

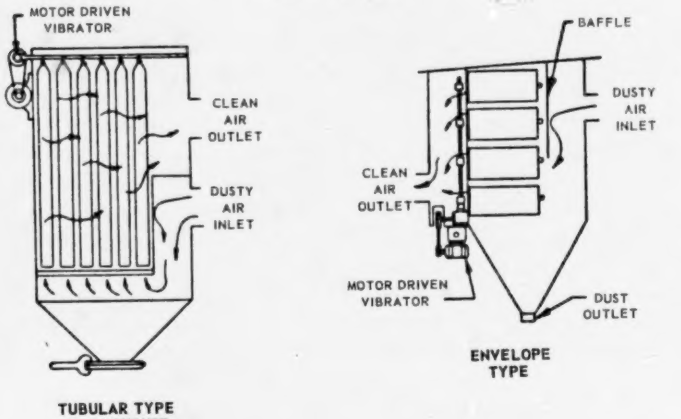
Filters

For the collection of extremely fine particulates where moderate conditions of temperature, humidity, and corrosion can be maintained, cloth filters have found wide application rating second only to cyclonic collectors in scope of application for industrial air pollution control. Fundamentally this type of collector is designed to remove solid dispersoids from the carrier gas by " filtration " of the gas through a porous medium. Two basic types of filter are at present employed; the first utilizes a fibrous medium as the collecting element, and the second utilizes the medium as the support for a layer of collected particles, relying upon this coat of the collected material to serve as the principal collecting element. The latter type is by far the more important for industrial application and normally utilizes a fabric as the supporting medium.

The most common type of fabric collector is the tubular type consisting of a structure in which cylindrical fabric bags are suspended vertically over a tube sheet with the open end of the bag attached to the tube sheet. The

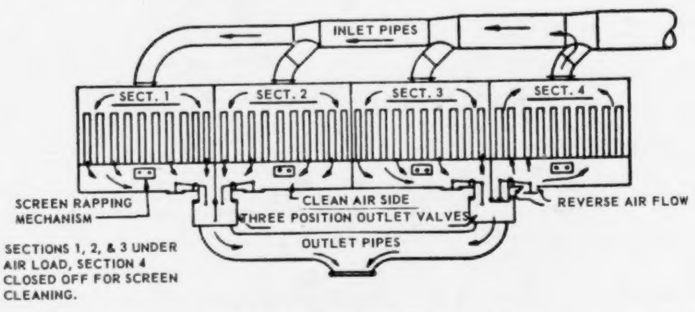
FIG. 3

FABRIC FILTERS


MOTOR DRIVEN VIBRATOR
CLEAN AIR OUTLET
DUSTY AIR INLET
BAFFLE
DUSTY AIR INLET
CLEAN AIR OUTLET
MOTOR DRIVEN VIBRATOR
DUST OUTLET

ENVELOPE TYPE

TUBULAR TYPE OR BAGHOUSE


INLET PIPES
SECT. 1
SECT. 2
SECT. 3
SECT. 4
SCREEN RAPPING MECHANISM
CLEAN AIR SIDE
THREE POSITION OUTLET VALVES
REVERSE AIR FLOW
OUTLET PIPES
SECTIONS 1, 2, & 3 UNDER AIR LOAD, SECTION 4 CLOSED OFF FOR SCREEN CLEANING.

MULTIPLE SECTION CONTINUOUS AUTOMATIC

tube sheet, in turn, forms the upper side of a gas entrance plenum which also serves as a dust collecting hopper. The particle-laden gases pass into the gas entrance plenum or expansion chamber. Impingement on a baffle plate and a reversal of gas direction cause the heavier particles to drop by gravity into the hopper. The carrier gas then flows upward into the tubes and thence outward through the fabric leaving the particulate matter as a " cake " on the inside of the bags. The filter bags are periodically cleaned by some combination of rapping, shaking, or vibration, or by reverse air flow, causing the filter cake to be loosened and to fall into the hopper below.

Many types of fabric filter are classified under the general category of " bag filter " (Fig. 3). Basically they differ only with respect to bag shape, type of housing, fan location, and method of cleaning the fabric.

Separation of the dispersoid from the carrier gas stream is not a simple filtration or sieving since the pores of the fabric employed in fabric filters are normally many times the size of the particles separated. The theory for removal of dispersoids by a fabric filter has not been fully developed. It is postulated that initial deposition of the dispersoid, prior to the formation of a pre-coat, takes place through interception and impingement of the particulates on the fabric fibres, and on account of Brownian diffusion, electrostatic attraction, and gravity settling within the pores Efficiencies during pre-coat formation are usually low but increase as the pre-coat is formed, until a final efficiency of separation, usually well over 99 %, is obtained. Once formed, this pre-coat becomes part of the filtering medium and the significant filtration mechanism then becomes sieving. In industrial applications with dust loadings normally encountered, this pre-coat usually builds up in a matter of minutes, sometimes even seconds. As previously indicated, the theory, as yet, is not exact as to the importance of each mechanism in the over-all separation, but the particle-size range over which each is considered to be effective is shown in Table 2.

TABLE 2

FILTRATION MECHANISMS AND PARTICLE SIZE RANGE OVER WHICH EACH IS EFFECTIVE

Mechanism	Particle-size range (μ)
Brownian diffusion	$<$ 0.01 - 0.2
Interruption	$>$ 1
Impingement	$>$ 2
Electrostatic deposition	$>$ 0.01
Sieving	$>$ Filter medium interstices

Source: Silverman, 1950.

Recent developments in the application of fabric filters for dispersoid control have been notable. Not only is there continued research toward the understanding of the mechanisms of filtration, but there is an increased scope of employment of this type of control equipment engendered by the development of a wide variety of filter media and by the utilization of gas cooling techniques in conjunction with the control systems.

Recognition of the factors having an adverse effect on dispersoid removal efficiency has resulted in improved design and application of fabric filters. Some of the more common design deficiencies are: (*a*) excessive filter ratios; (*b*) improper selection of filter media; and (*c*) lack of on-stream capacity during cleaning.

The term " filter ratio " is defined as the ratio of carrier gas volume to gross filter area, and is sometimes termed superficial face velocity. Excessive filter ratios have several deleterious effects on dispersoid removal efficiency. In practice, the employment of high filter ratios for fine particles such as metallurgical fume results in a rapid build-up of filter resistance. This, in turn, requires more frequent shaking of the bag and results in increased bag wear. Further, higher filter velocities, with corresponding increase in filter resistance, may reach levels inconsistent with reasonable power consumption. The effects of high filter velocity are amplified in the filtration of fine particles, but are much less pronounced in the filtration of fibrous dusts, which tend to form porous rather than packed cakes. Effects similar to those produced by excessive filter velocities are produced by high dispersoid concentrations in the gas stream. Decreased filter ratios are therefore recommended for high concentrations of dispersoids.

In practice, filter ratios range from 1 to 6 cubic feet[1] per minute per square foot[2] of cloth area; filter ratios of 3 cubic feet per minute per square foot of cloth area have been established as a common standard for normal dusts; however, the recommended ratio for fine dust and metallurgical fumes does not exceed ½-1 cubic foot per minute per square foot of cloth area.

Filter resistances corresponding to these filter ratios normally lie between 2 and 6 inches of water but some instances of satisfactory operation have been recorded with pressure drops as high as 12 inches of water. Total filter resistance consists of (*a*) that imposed by the fabric itself, and (*b*) that imposed by the collected dust layer formed on the surface of the fabric. Since flow through the filtering medium under conditions encountered in application is in the region of streamline flow, this total pressure drop can be expressed as a sum of two resistances, each of which is proportional to the filter velocity and to the effective thickness of the fabric and cake respectively.

$$\triangle p = K_o V_s + K_1 V_s W \qquad \text{(special units)} \qquad (5)$$

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[1] 1 cu.ft = 0.028 m³.

[2] 1 sq.ft = 0.092 m².

where, $\triangle p$ = pressure drop, inches of water
K_o = coefficient of fabric resistance, inches of water/(ft/min.)
V_s = filter ratio, (cu.ft/min.)/(sq.ft)
K_1 = coefficient of cake resistance, inches of water/(lb dust/sq.ft) (ft/min.)
W = weight of cake per unit cloth area, lb/sq.ft

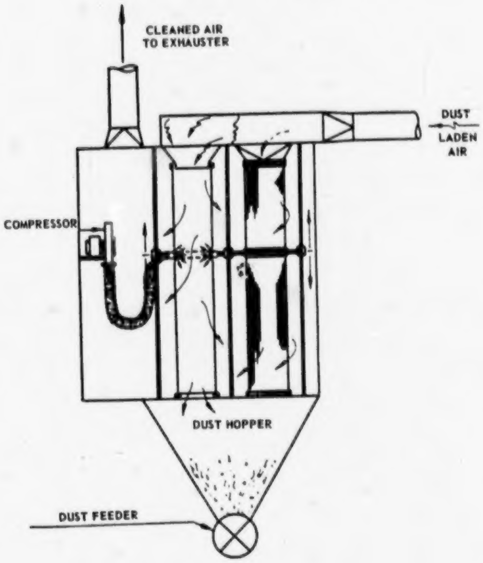
Improper application of filter media may result in unsatisfactory filter performance and in increased costs as a result of frequent bag replacement. In this regard, the fault normally lies in an inadequate appraisal of the operating limitations of the fabric. The more important of these limitations are temperature resistance, resistance to chemical attack, and abrasion resistance.

In the single compartment bag filter it is necessary that the gas flow to the unit be stopped during the cleaning or shaking cycle to allow the cake dislodged from the surface of the bag to fall by gravity to the hopper below. If requirements of the process being controlled are such that continuous operation is necessary, the bag filter must be of the multi-compartmented type to allow individual units of the bag filter to be successively off-stream during shaking. This is accomplished either manually in small units or by automatic programming control in large, fully automatic units. In this case sufficient cloth area must be provided to insure that filtering capacity will not be reduced during shaking periods when any one unit of the filter is off-stream. In this regard, it also should be noted that design should be based upon the maximum pressure drop anticipated, since pressure drop does not remain constant throughout a filtration cycle but varies from a minimum to a maximum value as filtration progresses with corresponding fluctuations in volumetric throughput.

A modification of the simple tubular type fabric collector consists of a unit in which cloth envelopes supported on wire frames are employed in place of the tubular bags. In contrast with the bag filter, air is introduced on the outside of each envelope and passes through the fabric into the frame of the unit and thence out of the collector. The filter fabric on this type equipment is cleaned normally by a rapping action on the supporting frame. For very fine or tacky dispersoids this type of unit is not normally applicable, since the efficiency of cleaning in such equipment is not as effective as that in the tubular type unit.

A comparatively recent innovation in the field of fabric filtration is the reverse jet filter (Fig. 4). This unit differs basically from the tubular type unit as to the method of cleaning and as to the type of filter medium employed. The tubular filters in this unit are cleaned by a high velocity air jet discharged from the inner side of a traversing annular ring which moves on the outside of the filter tube. The air jet passes through the fabric in a direction reverse to the normal flow and removes cake continuously

FIG. 4
REVERSE JET FILTER



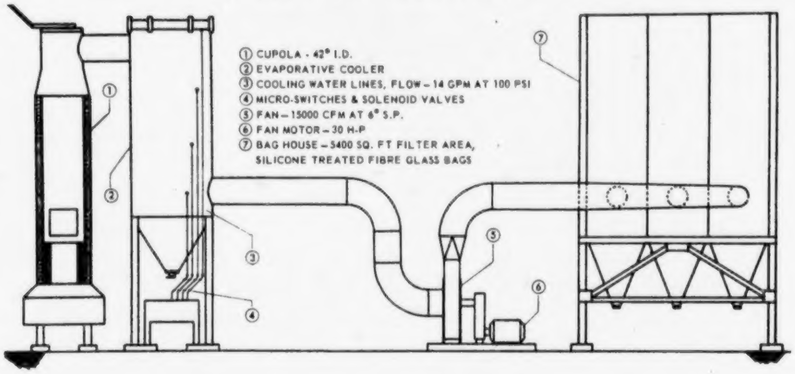
from the filter surface. Current designs employ pressed felt tubes as the filtration medium. Three advantages are apparent in this type of unit: (*a*) the unit requires no shut-downs or programming for cleaning; (*b*) it operates at filter ratios from 10 to 30 as opposed to the normal filter ratios of 1 to 6; and (*c*) the over-all filter resistance and, as a consequence, the volumetric throughput, can be maintained at a nearly constant value.

Fabric collectors have several distinct advantages which enhance their employment. These are: (*a*) high collection efficiencies for all particle sizes; (*b*) relatively constant collection efficiencies for variable gas flow rates and dispersoid concentrations; (*c*) simple construction; (*d*) dry disposal of the collected material; and (*e*) nominal power consumption. The use of fabric as a filtering medium, however, imposes several disadvantages: (*a*) operating limitations are imposed by high carrier gas temperatures, high carrier gas humidity (temperature of the gas must not fall below its dew point), and chemical activity of the gas or the dispersoid; (*b*) only solid particulates may be collected; (*c*) high maintenance and fabric replacement costs; and (*d*) large size of the equipment. The first of these limitations has been partly overcome through the use of gas cooling apparatus where the problem of high gas temperature exists. In recent years the development of fabrics capable of withstanding higher temperatures has made the employment of gas cooling or conditioning more practicable, since the degree of cooling required prior to their development often made gas conditioning economically unfeasible.

Three basic methods of lowering the temperature of gas streams have been employed: (*a*) cooling by radiation and convection; (*b*) cooling by evaporation; and (*c*) cooling by dilution with ambient air. The dilution method has not to date been used independently but rather as an adjunct to one of the first two methods. Radiation and convection cooling generally requires a greater initial capital outlay than evaporative cooling, but operation is less costly. Evaporative coolers (Fig. 5), however, necessitate the continuous use of cooling water with increased maintenance costs resulting from corrosion.

FIG. 5

EVAPORATIVE COOLER AND CLOTH FILTERING SYSTEM FOR A GRAY IRON CUPOLA

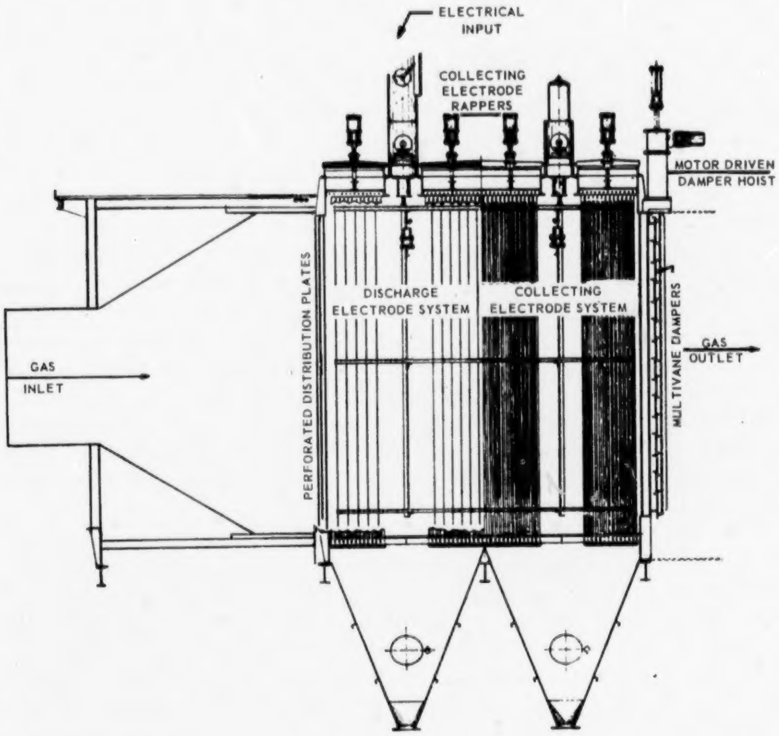

① CUPOLA - 42" I.D.
② EVAPORATIVE COOLER
③ COOLING WATER LINES, FLOW – 14 GPM AT 100 PSI
④ MICRO-SWITCHES & SOLENOID VALVES
⑤ FAN – 15000 CFM AT 6" S.P.
⑥ FAN MOTOR – 30 H-P
⑦ BAG HOUSE – 5400 SQ. FT FILTER AREA, SILICONE TREATED FIBRE GLASS BAGS
①
②
③
④
⑤
⑥
⑦

Electrical precipitators

Industrial air pollution control applications requiring high collection efficiencies through wide ranges of particle size and having extreme operating conditions such that heavy duty control equipment is necessary have best been served by electrical precipitation.

Basically, electrical precipitation is accomplished by passing the dispersoid-laden carrier gas between two electrodes across which a unidirectional, high voltage potential is impressed. One electrode, the discharge electrode, has a radius of curvature many times smaller than the second or collecting electrode. Owing to the differential in radius of curvature and to the high voltage potential impressed across the electrodes, a corona discharge is established in the region of the discharge electrode and, as a result, a powerful ionizing field is formed. Potentials as high as 100 000 volts are used. As the dispersoids in the carrier gas pass through this field, they become charged and migrate, under the action of the powerful electrical

FIG. 6
ELECTRICAL PRECIPITATOR


ELECTRICAL INPUT
COLLECTING ELECTRODE RAPPERS
MOTOR DRIVEN DAMPER HOIST
PERFORATED DISTRIBUTION PLATES
DISCHARGE ELECTRODE SYSTEM
COLLECTING ELECTRODE SYSTEM
MULTIVANE DAMPERS
GAS INLET
GAS OUTLET

field existing between the electrodes, to the oppositely charged collecting electrode. The particles, once deposited on the collecting electrode, lo[illegible] their charge and are removed mechanically by rapping, vibration, or washing to a hopper below.

Electrical precipitation as employed in air pollution control work is generally carried out in a single-stage precipitator in which gas ionization and particulate collection are combined into a single step (Fig. 6). There has been a very limited employment of a second type of precipitator, the two-stage unit, in which ionization is achieved in the first element of the equipment, followed by collection in the second element. Variations in design in each type differ essentially only in the details by which the following fundamental functions are accomplished: (*a*) gas ionization resulting from corona discharge; (*b*) charging of the particles; (*c*) migration of the particles toward the collecting electrode; (*d*) deposition of the particles on to the collecting electrode; and (*e*) removal of the collected material from the equipment.

There are two basic designs of the single-stage precipitator, the plate-type precipitator and the pipe-type precipitator. In the plate-type unit the collecting electrodes consist of parallel plates, either solid or of expanded metal, closely spaced rows of rods, chains, or wire, or specially formed shapes. In the pipe-type precipitator the collecting electrodes are formed by a nest of parallel tubes which may be square, round, or octagonal. In each case the discharge electrodes are wires or small, twisted rods which are suspended midway between the parallel collecting electrodes of the plate-type unit or axially along the length of the collecting electrodes in the pipe-type precipitator. Gas flow in the case of plate precipitators may be either parallel or perpendicular to the discharge electrodes and, of course, parallel to the collecting electrodes, but in the case of the pipe-type precipitator it must be parallel to the discharge electrodes. The corona discharge in both types of precipitator is maintained throughout the length of the precipitator, and it functions to provide ionization of the gas and to prevent redispersion of the precipitated dust by recharging neutral re-entrained particles. The discharge electrodes are usually suspended vertically from an insulated support and are kept taut by a weight at the bottom or by being stretched across an insulated frame.

Cleaning of the collecting electrodes for dry collection is accomplished by either periodic or continuous mechanical or electrical rapping to dislodge the collected material from the electrodes. In the film-cleaned precipitator, liquid is circulated over the collecting surfaces of the electrodes, and the material deposited is removed by washing. For liquid dispersoid collection the collected liquid is allowed to drain from the electrode surfaces under the action of gravity. It is common practice for the cleaning process to be carried out with the gas flow continuing and with the electrodes remaining energized, but this results in a tendency for re-entrainment of the dust during the cleaning cycle, with a consequent loss to the atmosphere. Specially formed collecting electrodes such as the hollow, pocket, or tulip types are used to reduce this tendency toward re-entrainment during rapping. Collecting electrodes in the plate-type unit generally are from 3 to 6 feet wide and from 10 to 18 feet high, and in the pipe-type precipitator from 6 to 15 feet long. Discharge electrode to collecting electrode spacing in both types usually ranges from 3 to 8 inches. Generally, the pipe-type precipitator is used for the removal of liquid dispersoids and the plate-type is employed for the collection of solid dispersoids.

In single-stage precipitators wherein particle charging and deposition take place simultaneously, the particles are not charged instantaneously; they assume a charge dependent upon electrostatic potential gradient, the dielectric constant, and the particle diameter (Deutsch, 1922, 1931a, b; Ladenburg, 1930; Mierdel, 1932). Velocity of migration of a particle toward the collecting electrode is a function of the electrical force acting on the particle and the resistance to movement of that particle due to gas friction. However, practical migration velocities are in general greater than those calculated, because of the action of electric wind (Mierdel, 1932). On this basis the following equation for collection efficiency has been derived (Deutsch, 1922, 1931a, b):

$$E_e = 1 - e^{-K_e \mu_e} \qquad (6)$$

where, E_e = weight collection efficiency, dimensionless
K_e = electrical precipitation constant, sec./cm or sec./ft
μ_e = migration velocity, cm/sec. or ft/sec.

The electrical precipitation constant is a function of the precipitator geometry and the gas velocity through the precipitator. Theoretically this equation for collection efficiency is applicable only for a given size particle, and over-all efficiency must, therefore, be obtained by an integration procedure for a specific particle-size distribution. In practice, however, particle migration velocity is generally determined experimentally for a class of particulates employing a pilot-scale precipitator, and the average experimental velocity thus determined is then applied in design calculations in the above equation. Further, from the equation it is seen that efficiency varies logarithmically with the precipitation constant and migration velocity; hence, size of a precipitator increases exponentially with increase in required efficiency.

Although a complete discussion of electrical precipitation is beyond the scope of this paper, it should be noted that there are a number of important practical problems involved in the design and application of this equipment. Many of these problems result from an inability to predict the exact behaviour of the particulate matter. In most cases this material is heterogeneous in size, shape and physical properties.

Electrical resistivity of the particulates has such a great effect on efficiency that a small change will cause an appreciable difference in the operating characteristics of the precipitator. High resistivity material, when deposited on the collecting electrode, will result not only in a reduction of potential across the gas stream, but also in back ionization with a consequent re-entrainment of the dust—both of which reduce efficiency (Beaver, 1946; Mierdel & Seeliger, 1936; Schmidt & Anderson, 1938). Research on this subject has indicated that the critical resistivity value for deposited dust layers is about 2×10^{10} ohm centimetres (Beaver, 1946).

Several approaches have been used to resolve this problem and all are based upon lowering the effective resistivity of the deposited layer.

Humidification, chemical gas conditioning, and temperature control have all been successfully employed. Conditioning agents include NH_3, NaCl, H_2SO_4, SO_3 and soluble sulfates.

Theoretically, corona discharge is continuous throughout the precipitator; practically, corona discharge emanates from a series of localized but mobile points on the discharge electrodes. Rectified alternating current is normally employed as the source of electrical energy and hence electrical field strength is not constant but varies continuously. High particulate concentrations increase the potential to produce corona and result in a reduction of current. These factors tend to bring about a reduction of efficiency but are compensated for in the experimental determination of particle migration velocities.

Depending upon the size, shape, and packing characteristics of the dispersoid, the design velocity through a precipitator may range from 3 to 10 feet per second. For any given set of conditions, the effect of increasing the velocity results in a reduction of retention time and in increased physical re-entrainment of the deposited material. Generally, there is a critical gas velocity for any specific dispersoid beyond which precipitation collection efficiency drops off rapidly. The problem of gas flow distribution in electrical precipitation is being given increased attention on account of the relationship which has been shown to exist between precipitation-zone gas velocity and collection efficiency. The assumption that average gas velocity is representative of actual conditions may in many instances be entirely erroneous. Practically, if the gas velocity profile is decidedly irregular, the expected efficiency of the precipitator may not be attained. Several methods for equal flow distribution are at present employed. These include simple enlargement sections, baffles, directional vanes, and perforated plates. In some applications such devices have been used both upstream and downstream from the precipitation zone.

Electrical precipitators are the most efficient type of control equipment for dispersoids in common use today. Advantages possessed by this type of equipment are: (*a*) high collection efficiency for all ranges of particle size and for high grain loadings under severe operating conditions; (*b*) dry disposal of solid particulates if desired; (*c*) low maintenance and operating costs; (*d*) low pressure drop; and (*e*) satisfactory handling of a large volume of high temperature gas. Disadvantages common to electrical precipitation include: (*a*) high initial cost; (*b*) large size of the equipment; and (*c*) possible explosion hazards.

For heavy duty operations, the disadvantage of high initial cost does not limit the application of precipitators, but for less demanding conditions, other high efficiency, less expensive, and less rugged equipment is normally employed. Collection efficiencies of 95% and higher are obtained with electrical precipitators in cases where particle sizes are as small as 0.01 μ and where operating conditions are extremely severe.

Scrubbers

In its most general sense the category of liquid scrubbers would include the various types of gas absorption equipment. As applicable, however, in the field of air pollution control the term " scrubber " usually is restricted to those devices which utilize a liquid to achieve or assist in the removal of solid or liquid dispersoids from the carrier gas stream. Generally, water is utilized as the scrubbing liquid, although in special cases other liquids have been employed.

Scrubbers are constructed employing such a multiplicity of design that no single type is representative of the category as a whole. Some units consist simply of an existing dry-type collector modified by the introduction of a liquid phase to assist in particulate matter removal and to prevent re-entrainment; other units are specifically designed to operate as wet collectors. In general, however, it may be said that dispersoids are collected in scrubbers by one or a combination of the following mechanisms: (*a*) impingement of the dispersoid on the liquid medium; (*b*) diffusion of the dispersoid on to the liquid medium; (*c*) condensation of the liquid medium vapours on the dispersoid, thereby increasing its size and weight; and (*d*) partitioning of the gas into extremely small elements to allow collection of the dispersoid by Brownian diffusion and gravitational settling on the gas-liquid interface.

The objective of dispersoid-liquid contact sections in the several types of scrubber is to present a large liquid surface area for the contact of the dispersoid in the carrier gas stream with the liquid phase. Both the mechanisms of impingement and of diffusion are favoured by high specific surface areas. Target efficiency for particles in the size range where Stokes' law applies is a function of the dimensionless group shown in the section on inertial separators. A general equation for impingement collection efficiency may be written (Johnstone & Roberts, 1949; Kleinschmidt, 1939):

$$E_s = 1 - e^{-(3E_t\,xy/2D_l)} \qquad (7)$$

where, E_s = weight collection efficiency, dimensionless
E_t = target efficiency, dimensionless
x = distance normal to gas flow through which collecting drop travels, cm or ft
y = ratio of volumetric liquid rate to volumetric gas rate, dimensionless
D_l = diameter of collecting drop, cm or ft

Impingement-type scrubbers, therefore, should operate more efficiently as the liquid droplet size is decreased. In consideration of the mechanism of impingement alone, however, it can be reasoned that because impingement efficiency depends upon relative velocity between dispersoid and liquid droplet, a minimum droplet size for optimum collection must exist. This is true because of the rapid acceleration to gas velocity undergone by very small droplets. It can be shown that impingement efficiencies fall off when collecting droplets smaller than 30-50 μ are employed.

In the extremely small dispersoid size range, the mechanism of diffusion becomes important in the collection of the dispersoids. Deposition on liquid droplets in the carrier gas stream will be effected by Brownian and eddy diffusion of the dispersoid. It is believed that the diffusion mechanism is predominant in the collection of submicron particles and may be sizable for dispersoids up to 5 μ in diameter. As in all diffusional processes the rate of diffusion is favoured by large areas for diffusion, thereby making small liquid droplets with high surface-to-volume ratios a necessity for high collection efficiencies. In most practical applications the dispersoid to be removed is heterogeneous and both impingement and diffusion mechanisms operate.

The mechanism of condensation occurs when cooling, resulting from the liquid spray, causes the dispersoid-laden carrier gas to pass through its dew point. Condensation will then take place with the submicron particles acting as condensation nuclei, thus increasing the effective size of the dispersoid. This mechanism is effective when initially hot gases containing relatively small dispersoid concentrations are conditioned.

Gas partitioning is achieved by dispersing the gas in very small increments through a liquid so that the distance between the suspended particles and the surrounding liquid-gas interface is extremely small. Separation of the dispersoid is then achieved by deposition on the liquid-gas interface by means of Brownian diffusion or gravitational settling. Effectiveness of this mechanism of separation increases with decreasing size of the gas increments.

The addition of surface-active agents to the liquid phase of scrubbers to enhance wetting characteristics is believed only to prevent re-entrainment of the deposited dust particles and not to enhance materially the actual process of dust deposition. Sufficient data on this, however, are not available and further study will be required before definite conclusions as to the effectiveness of this approach can be determined.

Various designs have been employed as scrubbers. A general list of the basic types would include spray towers, jet scrubbers, venturi scrubbers, cyclonic scrubbers, inertial scrubbers, mechanical scrubbers, and packed scrubbers.

The spray tower is a common type in which the dispersoid-laden carrier gas passes through banks of sprays, directed either parallel or normal to the gas stream to achieve liquid contact with the dispersoid. These sprays are followed by a set of staggered eliminator plates to remove the entrained water-dispersoid droplets. Water consumption varies from ½ to 2 gallons[1] of water per 1000 cubic feet of gas and pressure drops vary between 0.1 and 0.5 inch of water.

A modification of the spray tower is the water jet scrubber in which a high velocity jet of water is directed axially into the throat of a venturi section, effecting intimate liquid contact with the dispersoid-laden gas. This type of unit is normally followed by a simple gas reversal chamber to remove the entrained water-dispersoid droplets. Water consumption normally varies from 50 to 100 gallons per 1000 cubic feet of gas and the jet is capable of developing pressure gains up to 8 inches of water.

A recent development employing the venturi principle is the venturi scrubber in which the dispersoid-laden gas stream is directed through the venturi with a throat velocity of from 200 to 300 feet per second. Water sprays are introduced just preceding the venturi throat. The water and water-dispersoid droplets are then removed from the gas stream in a cyclonic spray separator. Water consumption varies from 3 to 10 gallons per 1000 cubic feet of a gas with a pressure drop across the entire unit of 10-15 inches of water.

A modification of a type of dry collector by the addition of a liquid phase is the cyclonic scrubber. In this unit a series of radial sprays are introduced into a typical cyclone. These sprays assist in the collection of the dispersoid and tend to prevent re-entrainment. Pressure drops usually vary from 2 to 8 inches of water with water consumptions varying from 3 to 10 gallons per 1000 cubic feet of gas.

Scrubbers in which liquid contact of the dispersoid is obtained as a result of the gas velocity itself are classified as inertial-type scrubbers. Two fundamental types fall within this classification. First, there is the impaction-type scrubber in which the liquid phase and the dispersoid-laden gases are intimately mixed and are then impacted on a plate to promote additional dispersoid-liquid contact. Impaction velocities may range from 30 to 150 feet per second, with pressure drops up to 30 inches of water. Secondly, there is the deflection-type scrubber, which utilizes deflector plates to effect liquid-dispersoid contact and to assist in the dispersion of the spray droplets. Water consumption ranges from 6 to 10 gallons per 1000 cubic feet of gas, and pressure drops may be as high as 6 inches.

In mechanical scrubbers (Fig. 7) liquid-dispersoid contact is achieved by the simultaneous introduction of the liquid medium and the gas stream on to rotating disks, blades, or perforated plates. In some cases stationary members are alternated with the rotating elements. Generally, high collection efficiencies for small particle sizes require high power consumption.

The final classification is the packed scrubber (Fig. 8). This unit is simply a conventional packed tower employing Raschig rings, Berl saddles, fibreglass, or other packing. Normally, gas flow is counter-current to liquid

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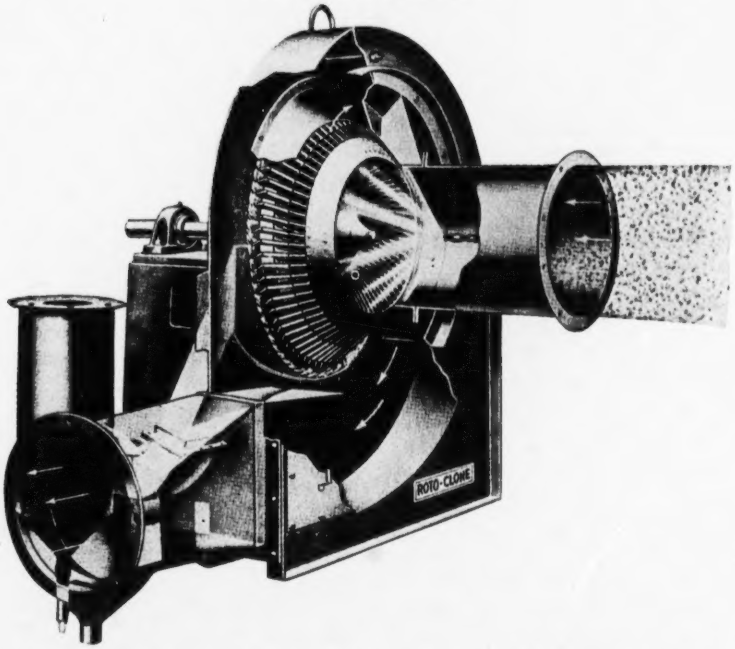
[1] 1 US gallon = 3.8 l.

FIG. 7
MECHANICAL SCRUBBER

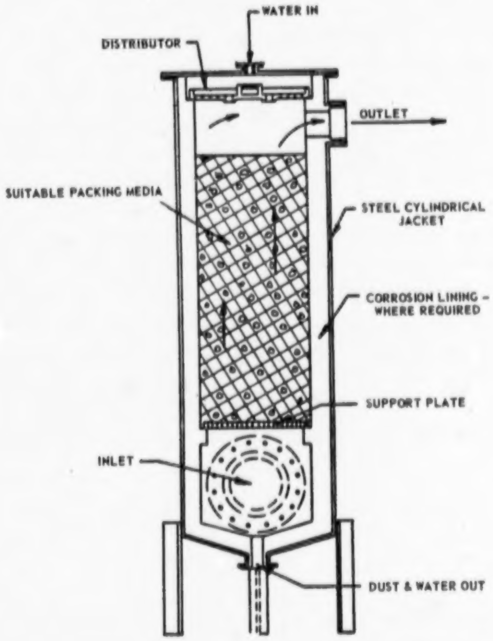


flow in this type of equipment. The separating mechanism is believed to be primarily impingement of the dispersoid on the packing itself with the liquid medium merely acting to clean continually the surfaces of the packing material. Excessive superficial tower velocities tend to result in channelling through the packing material with a consequent reduction in efficiency. Pressure drops are of the order of ½-10 inches of water.

Regardless of the scrubbing mechanism involved, power consumption is the critical factor determining collection efficiency. Energy input is consumed either by high-pressure atomization of the liquid medium, high velocity jetting of the liquid medium, high velocity of the dispersoid-laden carrier gases, or by mechanical impaction. Energy consumption in all cases will be approximately equal in attaining the same collection efficiencies. Simpler types of scrubber with lower energy inputs are effective in collectgin particles above 5-10 μ in diameter, while the more efficient, high energy input scrubbers will perform efficiently for collection of particles as small as 1-2 μ in diameter.

Since the types of scrubber available are so very diversified, the advantages and limitations of this classification of control equipment are difficult to generalize. It may be said, however, that this type of equipment has

FIG. 8
PACKED SCRUBBER


WATER IN
DISTRIBUTOR
OUTLET
SUITABLE PACKING MEDIA
STEEL CYLINDRICAL JACKET
CORROSION LINING - WHERE REQUIRED
SUPPORT PLATE
INLET
DUST & WATER OUT

the following advantages: (*a*) moderately high collection efficiencies below 10 μ; (*b*) moderate initial cost; and (*c*) applicability for high temperature installations. Scrubbers are limited in their application, however, in that they require: (*a*) wet disposal of the collected material; (*b*) variable power consumption, but high power consumption for higher efficiency; and (*c*) moderate to high maintenance costs, owing to corrosion and abrasion.

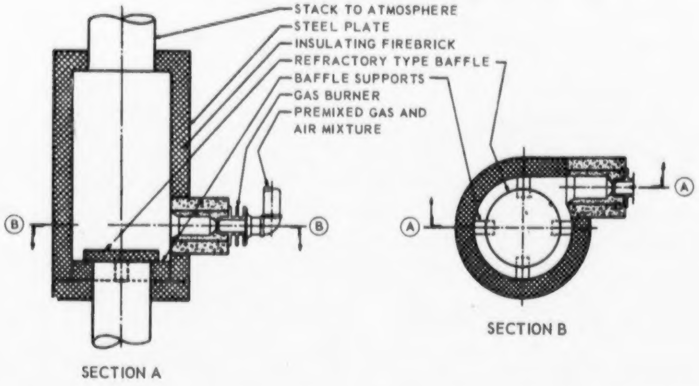
Control of gaseous contaminants

Control equipment for gaseous contaminants and odours must be considered apart from that for aerosols since the techniques employed differ basically. The control of noxious gases and odours is far from standardized mainly because of the widely varying chemical and physical properties and concentrations encountered. In addition, the importance attached to this type of contaminant is the result of a relatively recent trend in the air pollution field. Control equipment which has been used for gases and vapours includes combustion, absorption, and adsorption units.

Combustion is feasible where the pollutant gas or odorous components are oxidizable. Normally organic contaminants may be destroyed by exposure to temperatures in excess of 1200° F (650° C). Under certain conditions the use of catalysts may lower the required temperatures. The major limitation of this technique is that it may become too expensive when fuel values of the gaseous discharge are low, when moisture content of the discharge is high, or when exhaust volume is extremely large. Combustion-type control equipment has been used advantageously, however, in the petro-chemical, coffee roasting, paint and varnish, rendering, and fertilizer industries. Fundamentally these combustion units must provide: (*a*) sufficient combustion air for the oxidation reaction; (*b*) adequate temperature for continuous oxidation of the gaseous contaminant; and (*c*) adequate retention time in the high temperature combustion zone for completion of the oxidation reaction. Generally the incineration equipment consists of a single combustion chamber with provisions for maintaining continuous and intimate mixing of the contaminant-laden gas and the flame from the auxiliary burner (Fig. 9). The combustion chamber is so proportioned that the gas velocity and gas flow patterns established will produce adequate retention time in the combustion zone. Average retention time in such units is 0.2-0.3 seconds at temperatures of 1200° F and higher.

FIG. 9

WASTE GAS AND ODOUR INCINERATOR



Absorption involves the transfer of gas molecules into a liquid phase. It is a diffusional process and, as such, the driving force consists of concentration differentials between the bulk gas phase and the gas-liquid interface and between the interface and the bulk liquid. The various resistances to mass transfer should be as low as possible, and the absorption process is therefore favoured by high interfacial surface areas, turbulence in both fluid phases, and high diffusion coefficients. An absorbent is normally selected which has a high capacity to absorb without build-up of appreciable back pressures. To achieve this, the gas to be absorbed should have a high solubility in the chosen absorbent, or it should react irreversibly with the absorbing liquid.

Normally in industrial absorption operations the concentration of the solute gas in the carrier gas is relatively high. This may not be true where the control of air pollution is the objective. Very large gas volumes containing only low concentrations of pollutants are often handled and the equipment required may be quite large in relation to that employed in chemical processing. In addition, the carrier gas may contain particulate matter requiring that the gas absorption equipment also function as a wet dust collector.

In general, absorption equipment may be classified as spray chambers, mechanical contactors, bubble cap or sieve plate contactors, or packed towers. A spray chamber, as the name implies, is simply an empty chamber in which the gas stream is passed through curtains of liquid spray. Mechanical contactors employ mechanical agitation to accomplish intimate contact of gas and liquid. In a bubble cap or sieve plate contactor, the gas is passed upward through a series of plates on which pools of absorbent exist; bubble cap trays are used in the former and porous or perforated plates in the latter to support the liquid layers. Packed towers allow the liquid absorbent to flow by gravity downward through a bed of packing material while the gas stream moves either concurrently or counter-currently through the tower. In each of the above units, intimate gas-liquid contact is promoted over large interfacial areas.

The above types of contactors have certain relative advantages and disadvantages. Spray chambers have low pressure drops and will not plug; they also will function as simple spray-type dust collectors. In addition, these chambers have been found to be fairly effective in the removal of even rather insoluble gases and may, therefore, compare favourably with other types of equipment in certain applications (Pigford & Pyle, 1951; Sherwood & Pigford, 1952). Mechanical contactors are, of course, more complex in construction and higher in cost. They are quite useful, however, when the gas to be absorbed is relatively insoluble and liquid diffusion resistance is controlling. Packed towers may plug with solids, and cleaning is not easy. Consequently, grid packings rather than dumped packings are often used to overcome plugging tendencies and to lessen pressure drop. Plate towers will normally operate at higher pressure drops than packed towers but they may be much more easily cleaned and may be more effectively used as combination absorber dust collectors.

Adsorption is another diffusional process which is growing in importance as a separation mechanism. Adsorption is a surface phenomenon and requires the presence of large solid surface areas to be effective. In almost all cases these areas are internal as with porous material and may be almost unbelievably great per unit of adsorber volume. Adsorption may be primarily physical in nature or may include chemical surface reaction as in chemisorption.

Although adsorption has been used very successfully in certain industries for many years, the theory and techniques involved are not nearly so well known as those for distillation, filtration, extraction, etc. This is particularly true when adsorption is applied in a new field such as air pollution control. While considerable experimental work has been done to enable prediction of flow patterns, pressure drops, material balances, and transport and adsorption rates, there is not complete agreement on the various design procedures proposed. In actual practice the adsorptive capacity, height of bed, and stripping conditions for a given adsorption problem, in most cases, are obtained experimentally as a basis for design of full-scale equipment.

It appears that adsorption as a means of air pollution control may offer certain advantages for some classes of contaminant. These classifications include gases or vapours for which other means of collection are uneconomical, hazardous, or impossible. For example, many flammable organic compounds may be recovered with relatively high efficiency by adsorption at concentrations safely below the lower explosive limits of the compound. Also, since adsorption may give very complete clean-up even when pollutant concentrations are very low, unusually toxic or odorous compounds may sometimes be removed by adsorption on activated carbon, etc. when all other practical methods fail.

Adsorption equipment is almost always the packed bed although fluidized beds can also be used. Power consumption is dependent on the height of the adsorbing bed, the adsorber pellet size, and the flow rate, and pressure drops may vary from a few inches of water upwards.

The generation of heat of adsorption in a bed may make necessary pre-cooling of the effluent gas in some cases. It must be noted that normally both adsorptive capacities and collection efficiencies fall off with higher temperatures. Installation costs for adsorbers are high but maintenance and operating costs are not excessive. Small-sized units will operate as efficiently as larger ones and many times the value of the recovered material will enhance economic feasibility.

The Application of Process Changes for the Prevention of Air Pollution

In controlling air pollution by process changes, the method to be employed is completely dependent upon the particular process involved; hence, no fixed set of rules can be universally applied. Selection of the control method must be based on: (*a*) determination of the types of contaminant creating an air pollution problem; (*b*) complete evaluation of the plant processes to ascertain the units or operational areas discharging the offending contaminants; and (*c*) determination of specific adequate control techniques for those contaminants.

Four basic approaches to the control of contaminants through process modification are available: (*a*) substitution of raw materials or fuels; (*b*) modification of the process itself; (*c*) modification or replacement of process equipment; or (*d*) changes in operational practices.

The substitution of raw materials or fuel has often been used successfully as a method of atmospheric pollution control when the contaminant being eliminated from the fuel or raw material is not essential to the process. For example, bauxite flux may be substituted for fluorspar in open hearth practice and powdered sulfur has been replaced as a flux in the manufacture of magnesium castings. Both changes result in decreased emission of air pollutants. Also, the use of low-volatile coals in place of high-volatile coals has proved most effective in eliminating smoke and soot in many commercial and industrial heating applications, and the substitution of low-sulfur fuels for high-sulfur fuels has reduced the SO_2 discharges quite appreciably.

In modifying a given process to effect a reduction in atmospheric pollution, a unit operation may be eliminated or modified, or other unit operations may be substituted or added. Elimination of atmospheric pollution caused by the manufacture of intermediate raw materials can be achieved by acquiring such components through purchase from an outside supplier; thus, the manufacturing process creating air pollution can be discontinued.

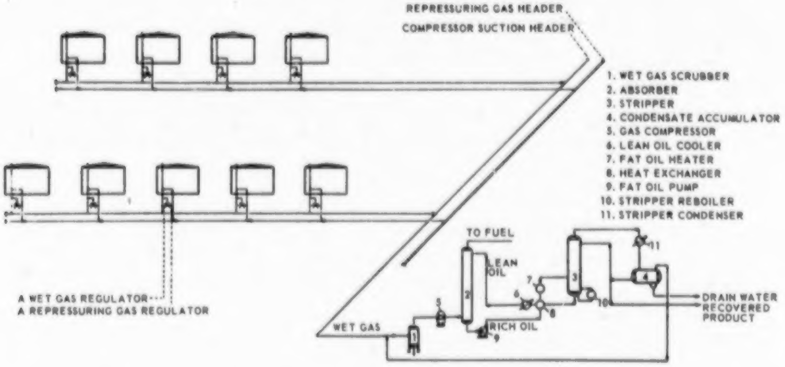
The addition of an operational step to reduce atmospheric pollution has been successful in brass foundry practice in which indirect fired furnaces are employed. A fluxing material is applied to the surface of the molten brass serving as an evaporation barrier and reducing the emission of brass fumes; this additional step is included strictly as an air pollution control measure. Also, the control of pollution by substituting one complete operation for another is illustrated in the disposal of combustible refuse where incineration may be discontinued in favour of sanitary land fill.

Changes in equipment as a method of reducing the release of contaminants to the atmosphere may include: (*a*) the modification of one or more items of basic process equipment; (*b*) the substitution of one type of equipment for another type; or (*c*) the replacement or repair of faulty or malfunctioning equipment. An illustration of this is the use of vapour recovery systems to control vapour losses in the handling of volatile materials (Fig. 10). Headers, compressors, absorbers, and condensers may be employed to return effluxing vapours to the system. The installation of sealed storage tanks—e.g., floating roof tanks—is another example (Fig. 11). In the cast iron foundry industry the substitution of reverberatory furnaces for cupolas has proved successful in reducing the contaminant discharge from this type of operation.

Changes of operational procedures for the sole purpose of reducing pollution discharge is well illustrated by the technique practised in power

FIG. 10

VAPOUR RECOVERY SYSTEM


REPRESSURING GAS HEADER
COMPRESSOR SUCTION HEADER
1. WET GAS SCRUBBER
2. ABSORBER
3. STRIPPER
4. CONDENSATE ACCUMULATOR
5. GAS COMPRESSOR
6. LEAN OIL COOLER
7. FAT OIL HEATER
8. HEAT EXCHANGER
9. FAT OIL PUMP
10. STRIPPER REBOILER
11. STRIPPER CONDENSER
TO FUEL
LEAN OIL
RICH OIL
WET GAS
DRAIN WATER
RECOVERED PRODUCT
A WET GAS REGULATOR
A REPRESSURING GAS REGULATOR
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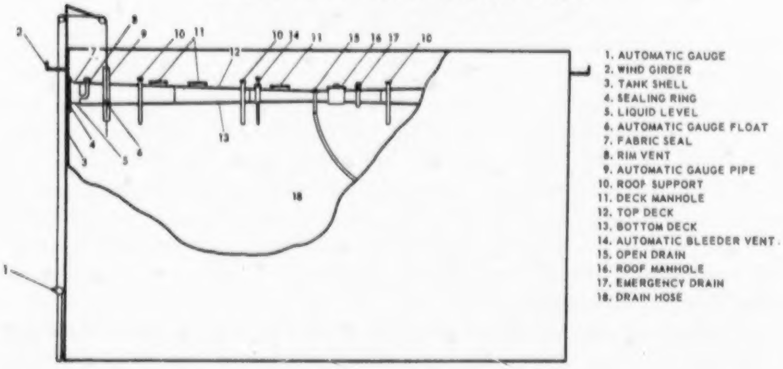
11

plant operation by the Tennessee Valley Authority. Here the rate of release of sulfurous emissions from coal burning operations is reduced during periods of adverse meteorological conditions by firing a low-sulfur fuel in place of the normal high-sulfur fuel. It is also common to reduce atmospheric contamination from boiler operation by employing recommended procedures for starting and cleaning fires and by using accepted stoking techniques, etc.

An important example of air pollution control by process changes lies in combustible waste disposal. This process will therefore be discussed in detail to illustrate the type of research which can be conducted for determining proper design and operational changes to reduce the emission of air pollutants.

FIG. 11

DOUBLE DECK FLOATING ROOF TANK


1. AUTOMATIC GAUGE
2. WIND GIRDER
3. TANK SHELL
4. SEALING RING
5. LIQUID LEVEL
6. AUTOMATIC GAUGE FLOAT
7. FABRIC SEAL
8. RIM VENT
9. AUTOMATIC GAUGE PIPE
10. ROOF SUPPORT
11. DECK MANHOLE
12. TOP DECK
13. BOTTOM DECK
14. AUTOMATIC BLEEDER VENT.
15. OPEN DRAIN
16. ROOF MANHOLE
17. EMERGENCY DRAIN
18. DRAIN HOSE
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1

Incineration of combustible wastes

A significant factor affecting the concentration of pollutants within the air mass of any specific area is the contaminant discharge produced from the disposal of combustible wastes by incineration. Other methods of disposal of such wastes include open dumping, sanitary land fill, and composting, none of which, when properly operated, creates any appreciable air pollution problem. However, the selection of a particular method of waste disposal must be based on such factors as the combustible content of the refuse, economic considerations relating to the various methods of disposal, and upon the sanitary and aesthetic requirements of the area.

Disposal of combustible wastes by incineration has the distinct advantage over other methods in that it reduces the volume of refuse by as much as 95 %, leaving a residue with little or no organic material. Such residues, therefore, require a minimum land usage for disposal and the disposal area is not subject to excessive settling after compaction.

Incineration is defined as any combustion process employed for the disposal of combustible wastes such as refuse, rubbish, garbage, animal remains, and solid, semi-solid, liquid, and gaseous waste products. To be effective from the air pollution standpoint, incineration must achieve: (*a*) maximum combustion efficiency in order to reduce atmospheric discharge of smoke, tars, malodorous compounds and other products of incomplete combustion, and (*b*) maximum retention of the incombustible solids in order to minimize fly ash and other solid discharges to the atmosphere.

Although engineering parameters are available for the design of industrial process furnaces burning fuels of uniform composition at specified rates, such data are not available for the design of incinerators. This results from the fact that incinerators are frequently batch fired and are required to perform satisfactorily over wide ranges of operating conditions while burning high-volatile fuels. Further, incinerators are normally charged at varying rates with heterogeneous materials of varying composition, and with corresponding variations in combustion air flow rate and air distribution. This results in widely fluctuating combustion chamber temperatures. Unless the incinerator is able to accommodate these wide ranges in operating characteristics, excessive atmospheric contaminants will be discharged.

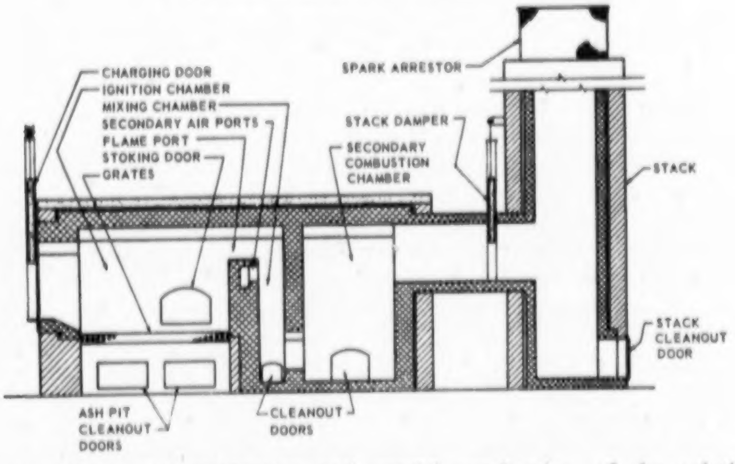
Incinerators may be classified broadly as to the number of combustion stages employed in the process—i.e. single stage or multiple stage. Single-chamber incineration equipment is so constructed that drying of the fuel, gasification and combustion of the volatile components of the fuel, and ignition and combustion of the fixed carbon proceed in a single stage. Combustion air is supplied both underfire and overfire, and mixing of the overfire air with the volatilized gases, necessary for good combustion, is comparatively poor owing to the configuration of the chamber and, of more import, to lack of facilities to expedite proper mixing.

Additional research to improve combustion efficiency in single-chamber incinerators is at present under way and preliminary results indicate that considerable improvement in this type of incineration equipment may be achieved (R. C. Corey, of the US Bureau of Mines, personal communication, June 1957). Tangential air jets are employed to improve mixing of the volatile unburned constituents of the fuel with combustion air. Burning rate and amount of excess combustion air have been indicated to be functions of both mass flow rate of air and Reynolds number of flow through the ports. Optimum conditions in a prototype experimental unit were obtained employing an average excess air of approximately 40 %. The height of the port above the fuel bed has also been shown to affect combustion efficiency and burning rate. Although these effects were not marked, in general combustion efficiency was greatest with higher ports and burning rate highest with low ports close to the fuel bed surface.

Proper designs for multiple-chamber incineration processes require that the equipment be so constructed as to promote a two-stage combustion mechanism (Fig. 12). Drying of the fuel, ignition and combustion of the fixed carbon, and gasification and partial combustion of the volatile components proceed in the ignition chamber or primary stage. The gas-phase combustion reaction is completed in the second stage, which consists of both a mixing chamber and a combustion chamber. Secondary air is admitted at the entrance to the mixing chamber to assist the completion of the gas-phase combustion reaction.

FIG. 12

LARGE MULTIPLE-CHAMBER INCINERATOR


CHARGING DOOR
IGNITION CHAMBER
MIXING CHAMBER
SECONDARY AIR PORTS
FLAME PORT
STOKING DOOR
GRATES
SPARK ARRESTOR
STACK DAMPER
SECONDARY COMBUSTION CHAMBER
STACK
STACK CLEANOUT DOOR
ASH PIT CLEANOUT DOORS
CLEANOUT DOORS

Three fundamental design concepts for multiple chamber incineration processes affect the combustion efficiency. These concepts are: (*a*) ratio of grate loading to combustion rate; (*b*) combustion air distribution; and (*c*) ignition chamber proportions.

Present data indicate that burning rates of from 40 to 50 pounds per square foot[1] of grate area per hour are optimum. It should be noted that these rates are applicable only for the large municipal-type incineration equipment, above 50 tons[2] per day capacity. For smaller capacity incinerators grate loading expressed in pounds per square foot of grate area per hour decreases as a logarithmic function of combustion rate expressed in pounds-per-unit time (Rose & Crabaugh, 1955):

$$G_L = 16.5 + 9 \log C_R \text{ (special units)} \quad (8)$$

where, G_L = optimum grate loading, (lb) / (sq.ft) (hr)
C_R = combustion rate, lb/min

The second concept fundamental to incinerator design when considered from the air pollution standpoint relates to combustion air distribution. Of significance in this respect is the ratio of underfire air (combustion air distributed through the grate) to overfire air. Present data indicate that optimum results are obtained when 65%-75% of the actual combustion air requirements for the incineration process, based on 100% excess air, are supplied overfire. The ratio must, however, be based upon stoichiometric air requirements for the particular chemical composition of the fuel concerned.

The third concept concerns ignition chamber proportions. In the past these have been considered to be related to ignition chamber volume. It has been found that a refinement of this concept exists in the relationship of arch height to grate area. While this relationship does establish the ignition chamber volume, it has been indicated to be more definitive in establishing combustion rate and contaminant discharge than direct volumetric relationships. An empirical formula for this relationship has been developed (Rose & Crabaugh, 1955):

$$A_H = \frac{6}{5} (G_a)^{0.364} \qquad \text{(special units)} \quad (9)$$

where, A_H = arch height, ft
G_a = grate area, sq.ft

Relatively little data are available on solid and gaseous discharge from incineration processes. Available data (Chass & Rose, 1953) indicate that commercial or industrial single-chamber incinerators with no adequate provisions for controlling combustion air, discharge an average of 20 pounds[1] of solid contaminants to the atmosphere per ton of material burned. Further, an average of 24 pounds of solids are discharged to the atmosphere for each ton of material consumed in a " backyard " type of incinerator and 19 pounds for every ton burned in a " chute fed " apartment house type of incinerator. Municipal incinerators of the multiple-chamber type employing secondary air and with provisions for combustion air control produce an average of 8.8 pounds of solids per ton of material burned, with a range of 3.4-17.8 pounds per ton. Unpublished data on industrial and commercial multiple-chamber incinerators, with capacities up to 4000 pounds per hour, indicate an average loss of solid contaminants to the atmosphere of 6.8 pounds per ton of material burned, with a range of 3.0-14.7 pounds per ton.

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[1] 1 lb/sq.ft = 4.882 kg/m².

[2] 1 short ton = 0.9 metric ton.

ACKNOWLEDGEMENT

Fig. 1-12 are reproduced by courtesy of the Los Angeles County Air Pollution Control District and the American Air Filter Company, Inc.

REFERENCES

Albrecht, F. (1931) *Z. Phys.*, **32**, 48

Alden, J. L. (1940) *Design of industrial exhaust systems*, New York

American Petroleum Institute (1955) *Cyclone dust collectors*, New York

Beaver, C. E. (1946) *Trans. Amer. Inst. chem. Engrs*, **42**, 251

Chass, R. L. & Rose, A. H. jr (1953) *Discharge from municipal incinerators*, Los Angeles (Los Angeles County Air Pollution Control District, Publication No. 44)

Deutsch, W. (1922) *Ann. Phys.*, **68**, 335

Deutsch, W. (1931a) *Ann. Phys.* **9**, 249

Deutsch, W. (1931b) *Ann. Phys.*, **10**, 847

Drijver, W. (1937) *Wärme (Berl.)*, **63**, 333

Johnstone, H. F & Roberts, M. H. (1949) *Industr. Engng Chem.*, **41**, 2417

Kleinschmidt, R. V. (1939) *Chem. metallurg. Engng*, **46**, 487

Ladenburg, R. (1930) *Ann. Phys.*, **4**, 863

Langmuir, I. & Blodgett, K. B. (1946) *U.S. Army Air Force Technical Report* No. 5418

Lapple, C. E. (1950) *Dust and mist collection.* In: Perry, J. H., ed. *Chemical engineers' handbook*, New York

Lapple, C. E. (1951) *Fluid and particle mechanics*, Newark, p. 306 (University of Delaware)

Linden, A. J. ter (1949) *Proc. Inst. mech. Engrs*, **160**, 233

Magill, P. L., Holden, F. R. & Ackley, C., ed. (1956) *Air pollution handbook*, New York

Mierdel, G. (1932) *Z. tech. Phys.*, **13**, 564

Mierdel, G. & Seeliger, R. (1936) *Trans. Faraday Soc.*, **32**, 1284

Pigford, R. L. & Pyle, C. (1951) *Industr. Engng Chem.*, **43**, 1649

Rose, A. H., jr & Crabaugh, H. R. (1955) *Research findings in standards of incinerator design.* In: Mallette, F. S., ed. *Problems and control of air pollution*, New York

Rosin, P., Rammler, E. & Intelmann, W. (1932) *Z. Ver. dtsch. Ing.*, **76**, 433

---

[1] 1 lb = 0.45 kg.

Schmidt, W. A. & Anderson, E. (1938) *Electr. Engr*, **57**, 332
Sell, W. (1931) *ForschArb. IngWes.*, **2**, Forschungsheft 347
Shepherd, C. B. & Lapple, C. E. (1939) *Industr. Engng Chem.*, **31**, 972
Shepherd, C. B. & Lapple, C. E. (1940) *Industr. Engng Chem.*, **32**, 1246
Sherwood, T. K. & Pigford, R. L. (1952) *Absorption and extraction*, 2nd ed., New York
Silverman, L. (1950) *Heat. & Ventilating*, **47**, 68 (reference section)
Van Tongeran, H. (1935) *Mech. Engr*, **57**, 753
Wellman, X. (1938) *Feuerungstechnik*, **26**, 137

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FUEL SELECTION AND UTILIZATION

Primary Energy Sources

The main natural sources upon which man depends for the energy which he requires for industrial production are coal and oil. The contribution from others, such as hydro-electricity, is in total small, although locally it may be of very great significance. With increasing industrialization, as well as with the mechanization of transport and agriculture, the demands for energy are on the verge of outstripping the capacity to supply them, conveniently and economically, from conventional resources.

It was where coal was readily and cheaply available, and other circumstances were favourable, that great centres of industry were first developed. The rapid growth of the petroleum industry during the last half-century is developing a challenge to coal as the pre-eminent fuel for industry. Not only has this led to the establishment of new industries close to the oilfields, in the same way as electro-metallurgical industry is located, if possible, where hydro-electricity is cheaply produced. But because oil is readily transported, it has become a competitor to coal in the older industrial centres.

The trend can be illustrated by statistics for a few of the leading industrial countries (Organisation for European Economic Co-operation, 1956; United States, President's Materials Policy Commission, 1952). However, the figures available are for the total energy consumption of each country, not of its industry alone, and it should be borne in mind that the major use of petroleum products is still for transport. In the USA, which possesses great resources of both oil and coal, over 90 % of the energy requirements in 1900 were derived from coal; by 1950 the proportion had been reduced to under one-half. Western Europe is comparatively poor in indigenous petroleum, but still a similar trend can be detected. At present, the United

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Kingdom derives over 85 % of its energy from coal, but this will be reduced, it is expected, to 75 % by 1965. Western Germany's total energy consumption is the second highest in Europe, and is still derived predominantly from coal; petroleum products amount to only 9 % of the total consumption at present. France, on the other hand, which must supplement its production of both coal and oil by imports, now derives 22 % of its energy from petroleum, 10 % from hydro-power, and the remaining 68 % from coal. Italy, poor in coal resources, has based much of its rising industrial production on petroleum, which now accounts for 42 % of its consumption, as compared with 30 % from hydro-power, and the remaining 28 % from coal.

The increasing use of oil as an industrial fuel has considerable bearing on atmospheric pollution. Smoke, grit and sulfur dioxide are the major sources of pollution from the combustion of fuel. While the combustion of both coal and oil can be controlled so as to prevent the emission of smoke, that control is generally easier with oil than with coal. If a further generalization be permitted, the danger of emitting smoke is greater from a small installation burning small quantities of coal than from a large one. With oil-burning the size effect is less marked, and it is therefore of importance that oil can more readily compete with coal as a fuel for small installations than for large ones. Grit emission is a problem peculiar to the use of solid fuel, but one for which technical solutions are available, as will be seen later. Sulfur dioxide, however, is encountered in the combustion gases from both fuels, and in greater amounts when oil is the fuel; the elimination of sulfur dioxide from flue gases is a costly operation, rarely practised.

To summarize then, the use of oil to supplement coal as a source of industrial energy reduces pollution from smoke and grit but increases somewhat the output of sulfur dioxide.

Secondary Energy Sources

Atmospheric pollution is also affected by the tendency for industry to supplement the energy it derives, within its own premises, from coal and oil by purchasing electricity and gas, which have been generated from the primary fuels at central stations and distributed by public utilities. While there is a substantial use of electricity for lighting, heating and for electrometallurgical purposes, the main reason why industry requires electricity is to provide motive power. The decision whether the power requirements of a factory should be met from its own installation or by purchase from the public utility is an intricate one, depending upon the size of the factory and the nature of the manufacturing processes undertaken, *inter alia*. In total, however, the amounts of electricity purchased far exceed, in most countries, those generated by industry on its own premises.

Manufactured gas is purchased for heating furnaces and other special processes, in which accurate control of temperature or cleanliness is of major importance, as well as for space-heating. While, strictly, natural gas should be classified as a primary energy source, it is conveniently included among the fuels distributed by supply companies, especially as the considerations which lead to its choice in the fortunate countries where it is available are those that dictate the purchase of manufactured gas elsewhere.

At their point of consumption, of course, gas and electricity do not pollute the atmosphere. At the central power station, pollution may arise, since the generation of electricity depends on the combustion of coal or oil. But since the units are large ones and subject to strict technical control, the incidence of pollution is, in fact, minimized. The process of gas manufacture need emit neither smoke, grit not sulfur into the atmosphere, although it is attended by special problems in dealing with liquid effluents. Of course, wherever large tonnages of solid fuel are handled, either at gas-works or power stations, there is a danger of dust dispersal, but again the large scale of operation permits use of the most effective means of preventing a nuisance arising.

In the manufacture of gas from coal, a solid residue, coke, is left. It may be either gasified separately, in the so-called water-gas process, or sold for domestic and industrial purposes. As its combustion takes place smokelessly, and can be carried out efficiently in small units, the availability of coke is of great importance in the avoidance of atmospheric pollution.

In the closely related coke-oven industry, the manufacture of coke, suitable for the reduction of iron-ore in blast furnaces, is the major objective. Gas in this case was formerly regarded as a by-product, but to an increasing extent it is now delivered into the distribution networks of the public utilities.

Coal, oil, electricity, gas and coke are, then, the major sources from which industry derives the energy and heat it requires. The list is not exhaustive, and indeed reference is made to some of the other secondary sources later in this paper. Potentially, the extent of atmospheric pollution from industrial sources is growing, not only because industrialization is spreading rapidly but because, within industry, machine-power is replacing man-power. Offsetting to some extent this potential growth is the changing pattern of fuel utilization, to which reference has already been made: first, the rising proportion of oil used to supplement or replace coal as a primary fuel; secondly, the trend within industry to purchase gas and electricity in increasing quantities.

Industrial Processes

From this general view of fuel utilization in its relation to atmospheric pollution, it is now necessary to turn to the particular, to the purposes for which fuel is used in the individual factory, and the measures adopted there to reduce pollution. However, these purposes are so multifarious that it is quite impracticable to consider them all, in their relation to atmospheric pollution, within a single chapter.

Drastic simplification is necessary and so the following procedure has been adopted. The largest single industrial use of fuel is in combustion under boilers, to provide for space-heating, process steam and sometimes for power generation. The consideration, in the following pages, of ways in which to reduce the emission of smoke, grit and sulfur dioxide respectively is primarily in terms of this major use. Many of the principles and some of the equipment described are applicable to other uses of fuel—e.g., in furnaces, cement manufacture, foundries and the ceramic industries—but special problems also arise there. These are briefly noted in a later section.

For a comprehensive treatment, the reader is referred to the *Proceedings of the conference on the mechanical engineer's contribution to clean air,*

TABLE 1

SUMMARY OF FUEL BURNT IN 1955 IN THE UNITED KINGDOM

Class of consumer	Fuel burnt (millions of long tons) [a]			Pollutants discharged (millions of long tons) [a]		
	Solid	Liquid	Coke-oven gas, unpurified	Smoke	Grit and dust [b]	SO_2
Coal:						
Domestic	38			0.9	0.1	0.9
Electricity works	43			small	0.3	1.2
Railways	13			0.3	0.1	0.4
Industrial and miscellaneous [c]	67		1.0	0.8	0.3	1.8
Coke ovens [d]			1.6	small	small	0.1
Gas-works	6 [e]			small	small	0.2
Coke:						
(Excluding consumption in gas-works and blast furnaces)	15			nil	small	0.4
Liquid fuels :						
Diesel and gas oil		3.8		small	nil	0.1
Fuel oil		5.4		small	nil	0.3
Creosote-pitch mixture		0.8			nil	small
Total	182	10.0	2.6	2.0	0.8	5.4

[a] 1 long ton = 1,016 metric tons.

[b] This column does not include grit and dust emissions, estimated at about 0,5 millions tons, from industrial processes other than steam raising.

[c] Includes general industry, collieries, patent fuel works, non-industrial establishments, waterworks and the Service Departments.

[d] Although, on average, the smoke and grit discharged from gas-works and coke ovens is small, the pollution from certain types of plant, and particularly from older coke-oven plants, can have a serious local concentration.

[e] Coke.

sponsored by the Institution of Mechanical Engineers and held in London from 19 to 21 February 1957. The following sections of this chapter, indeed, consist largely of a summary of contributions to that conference.

At this stage, it may be useful to have some measure of the extent of pollution which is caused by fuel-burning for different purposes. Table 1 summarizes the quantities of fuel consumed in the United Kingdom in 1955, and provides an estimate, which can of course be only a rough one, of the amounts of pollutants discharged. In one respect, the picture presented is radically different from conditions in other countries. The major emission of smoke is attributed to the domestic consumer, owing to the adherence of the British householder to the open coal fire. Pollution from the railways is another major source, but this will be steadily reduced during the next decade, as steam locomotives are replaced by Diesel and electric traction. With these exceptions, the incidence of pollution displayed in the table may be regarded as reasonably representative of what is to be encountered in a highly industrialized and densely populated community.

Prevention of Smoke Emission

Any fuel can be burnt without the emission of smoke, provided it has been chosen to suit the design of the appliance in which combustion takes place. There is considerable latitude in this matching of fuel and appliance, and generally the reason why smoke emission occurs is that the equipment has been badly maintained, or it is not being operated properly. The main principles that must be observed in the combustion of each fuel are outlined in the following paragraphs.

Oil

Oil in bulk is not very inflammable, and to burn it efficiently, it must be split up by pressure into small droplets in the combustion air. This process, called atomization, together with the effective mixing of the oil droplets with air, is controlled by the design of the oil-burner, a highly specialized branch of engineering. The degree of atomization is also greatly affected by the viscosity of the oil, and hence when the more viscous petroleum fractions are to be burnt, the fuel is preheated before delivery to the burner.

Generally, the flame is lit by an electrical ignition system. Thereafter radiation from the walls of the enclosure or from the burning oil is sufficient to ignite the fresh droplets as they emerge from the nozzle of the burner. The danger of smoke emission is greatest when starting up, or when the flame impinges on a cold surface. Refractory surfaces are therefore often incorporated in the combustion space to provide adequate radiation.

Finally, in order to prevent smoke emission, it is of course essential that the ratio of air to fuel should be controlled to provide a slight excess of air over that theoretically required for complete combustion. Provided these conditions are met and, as has already been emphasized, the standards of maintenance and operation have not been relaxed, oil is not a difficult fuel to burn smokelessly under steady load. Difficulty is more likely to be encountered in small installations which are lightly loaded--for example, in central-heating plants. Oil is well suited to these applications, because of the ease with which its combustion can be started and stopped. If there are long intervals between the firing cycles, however, care is needed to avoid the emission of smoke on re-ignition.

Coal tar

Crude coal tar is a by-product of the carbonization of coal at gas-works and coke-oven plants. It is distilled for the production of refined products, which include all types of coal tar fuels, varying in viscosity from fluid creosote to soft pitch. For practical purposes, the measures to be taken to ensure their smokeless combustion are the same as those which apply to the combustion of oil. If these precautions are not observed, the combustion of coal tar fuels may be attended by the formation of black smoke. Coal tar fuels generally contain a higher carbon content than petroleum fuels and produce hotter and more radiant flames.

Coal

Industrial coals are a heterogeneous mixture of coal substance and impurities, such as moisture and ash; they are supplied in a wide range of size grading. The coal substance itself (dry, ashless coal) varies widely in composition and properties, according to the coalfield, seam and pit from which it is mined. These variable properties include the chemical composition, the volatile content, and the caking properties, and they affect the combustion process considerably.

At modern power stations and in the larger industrial installations, coal is often pulverized before burning in conditions simulating the combustion of liquid fuels. Smoke production is then controllable but the prevention of grit emission becomes a major problem, as will be seen in the following section. Elsewhere the coal is delivered on to a grate, and its combustion takes place in two stages.

On the grate itself, the solid fuel is burnt by means of primary air passing through the fuel bed. The volatile matter driven off during this process is burnt by means of secondary air admitted over the fire. Good combustion requires good air distribution and, therefore, a fuel bed of uniform resistance to the passage of primary air.

Irregularities in the fuel bed result in inefficient combustion to an extent which depends primarily upon the properties of the fuel, but also varies with the combustion appliance. Among the coal properties affecting uniformity of the fuel bed are the following:

(1) The size grading is very important, particularly if the coal contains a high proportion of fines. With such coals, segregation almost invariably occurs in the fuel bed, with consequent maldistribution of the primary air flow. Bare patches occur in areas with low air resistance while areas of fine coal are starved of air. Conditioning of the fuel with water lessens the resistance of the fines and thus leads to better air distribution. The amount of water which may be added is sometimes limited by the feed mechanism of the stoker.

(2) The caking properties of a coal can affect the evenness of a fuel bed. With non-caking or weakly caking coals, the fuel bed structure and the air distribution are not changed during the combustion process, and each particle burns off separately. If, however, the coal has marked caking properties, the particles tend to bond together, forming lumps which burn slowly, whilst excess air is able to pass through the fissures which form between them. With strongly caking coals, the fuel bed may become very irregular and require frequent breaking up.

(3) Ash in fuel also affects the efficiency of combustion. If the ash has a high fusion temperature, it may prevent air from reaching the fuel and cause incomplete combustion. If the ash has a low fusion temperature, it will probably clinker and cause uneven air distribution through the fuel bed.

(4) The fines content of a coal, as already noted, may cause irregular fuel beds unless suitably conditioned, but may also have a bad effect on the operation of certain firing appliances.

Even this brief analysis may serve to indicate that the design of coal-burning equipment and the selection of the correct grade and rank of coal for any particular unit are an intricate technology. For examples of its application to particular conditions, reference may be made to the following papers: (Arthur & Napier, 1952; Arthur & Smith, 1956; Cosby, 1949; Dunningham, 1957; Gunn & Hawksley, 1957). Here it must be sufficient to note some general trends, which no doubt vary in significance in different parts of the world, according to local circumstances. In order to reduce labour costs and to increase combustion efficiency, the trend has for many years been to replace hand-firing by mechanical stokers, on boilers with ratings of 7000 lb. (3175 kg) of steam an hour, or even less. On balance, this has reduced atmospheric pollution by smoke, but sometimes it has increased grit emission.

Mechanical firing must result in some selectivity in choice of coal, and one trend has been to specify closely the fuel to be supplied for any particular appliance, or *vice versa*, in order to secure maximum efficiency of combustion. In those countries where the supply of coal is easy, this close specification is of course desirable. In other countries, of recent years, demand for coal has outstripped supply, and the mechanization of mining methods has led to the production of coal with higher ash contents and higher proportions of fines. Much thought has therefore been devoted to the development of firing appliances which will burn such coals without serious loss of efficiency. Clearly, supplies of coal which contain more ash and more fine particles enhance the danger of grit emission. This is a problem to be taken into account in the design of appliances, and attempts to solve it are resulting in the installation of grit arresters on small boiler plants, where formerly that would not have been considered necessary.

Coke

Coke is the residue remaining after the volatile matter has been driven from coal by heating in absence of air—the carbonization process. Two main types of coke are produced, metallurgical coke at coke-oven plants and gas coke at gas-works. The former is made from the best coking coals to ensure a coke of high mechanical strength for use in blast furnaces and other metallurgical furnaces. Gas coke on the other hand is widely distributed in some countries as an industrial and domestic fuel. It varies in its characteristics—but to a far less extent than coal—according to the carbonizing process used, the type of coal, its ash content and the carbonizing temperature employed. It normally contains about 90 % carbon, is comparatively friable and is much more reactive and readily ignitable than metallurgical coke.

Coke is a smokeless fuel, but with excessive draught grit emission may be severe, particularly when burning fines. With insufficient draught, the fire is liable to extinction. The main factor influencing the combustion characteristics of gas coke is size, and this must be considered when selecting a suitable coke-burning appliance.

Briquettes

Briquettes are manufactured from fine coal by agglomerating it into compact masses. This is usually done by mixing the fine coal with a binder, such as pitch, and compressing it in moulds which are heated sufficiently to melt the binder. Briquettes made in this manner, without being strongly heated, will retain the volatile content of the fine coal in addition to that of the added pitch, and are therefore liable to produce more smoke than the original coal. The considerations which apply to the smokeless combustion of coal apply in exactly the same way to uncarbonized briquettes.

Some smokeless briquettes are now being produced from low volatile coals, by carbonization at about 800° C. They will burn smokelessly without special precautions in a similar manner to coke.

Producer gas and water gas

Producer gas is an industrial fuel widely used in the metallurgical industries, in the manufacture of glass, refractories and ceramics, and for the heating of retorts or ovens in the carbonizing industries. It is made by passing a continuous mixture of air and steam through an incandescent bed of coal or coke. It is variable in composition but generally will consist of about 50%-55% nitrogen, 25%-30% carbon monoxide, 10%-12% hydrogen, and small quantities of other gases; it has a calorific value of from 110 to 155 B.Th.U./cu. ft/°F (approximately 1760-2480 Kcal/m^3/°C). In many applications, the hot gas is conveyed directly from the producer to the furnace in which it is to be burnt. It is obvious, from the nature of its production process, that the gas will be dirty, since it carries with it small particles of coke and ash from the fuel bed. It may also contain tar. Therefore, while producer gas is itself a smokeless fuel, unless it is cleaned it may be the cause of grit emission.

Water gas is not a general industrial fuel, being made normally only by gas-works and certain large chemical works. It consists almost entirely of carbon monoxide and hydrogen and, like producer gas, it is smokeless but requires cleaning.

Control of Grit and Dust Emission

The methods of controlling grit and dust emission can be classified as follows:

(*a*) dilution (use of chimneys)

(*b*) arresters (including inertial collectors, filters, precipitators and other devices for catching particulate matter)

(*c*) scrubbers

(*d*) sonic and ultrasonic agglomeration.

Dilution

Tall chimneys are of limited application, and are not a positive means of controlling pollution (Hawkins & Nonhebel, 1955). They should be considered as ancillary to other more positive methods, and not as sufficient by themselves. They can, of course, handle tremendous quantities of pollutant, and while their initial cost is high maintenance is quite low.

As a result of direct measurement and wind tunnel experiments, much is now known of the influence of chimney height on the dispersal of both solid and gaseous effluents. The following are some of the more important general points:

(1) The plume of gases emerging from the top of the chimney tends to expand at a solid angle of about 20° under normal wind conditions.

(2) Any building or obstruction adjacent to the chimney will cause down-draughts on the lee side, affecting air currents to a height some 1½-2½ times the height of the chimney. For isolated plants, therefore, the height of the chimney should be at least 2 ½ times that of surrounding buildings to ensure that the plume is not brought quickly down to ground level. It is obvious that the higher the chimney, the greater will be the dispersion and the smaller the concentration of " fall-out " at any point. For power stations and many industrial plants, chimney heights of 250-350 feet (about 75-100 m) and more are necessary.

(3) A chimney will itself cause down-draught eddies. These can be minimized by increasing the velocity of discharge and by suitable design of the chimney mouth.

(4) Theory and practice confirm that it is advantageous to combine, in a single chimney, the discharge from as many effluent-producing units as possible.

Arresters

Arresters comprise an extensive array of equipment but, in general, they can be classified into three sub-groups:

(*a*) inertial separators
(*b*) filters
(*c*) precipitators.

Inertial separators

These take two forms: centrifugal, e.g., cyclones, or trajectory, e.g., louver-type collectors. On the basis of cost per unit volume of gas handled they are probably the cheapest method of controlling particulate matter.

Cyclones [1] are widely used for industrial dust collection, either alone or followed by secondary collectors. The dust particles are subjected to an outward centrifugal force by admission of the dirty gases tangentially at the periphery of a cylindrical vessel. An inward viscous drag opposes the centrifugal force, and the balance of forces determines whether the particle will move to the wall and thence into the dust hopper or be carried inwards to the " clean-gas " exit.

The efficiency of a cyclone can be increased by irrigation of its walls, if a wet disposal system can be tolerated. Generally, the use of irrigation should be regarded as an expedient to improve the efficiency of an existing plant.

Stairmand & Kelsey (1955) suggest that cyclones should eliminate dust deposition nuisances from most small steam-raising plants, but have little effect on the appearance of the plume from the chimney.

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[1] A diagrammatic representation of a cyclone dust collector is shown on page 316 (" Prevention and Control of Air Pollution by Process Changes or Equipment, " by A. H. Rose, D. G. Stephan & R. L. Stenburg).

Filters

It is possible to obtain filters for handling almost any type of particle with any desired degree of efficiency, and they probably constitute the most effective means of controlling atmospheric pollutants. Their main drawbacks in combustion applications are that as the collection efficiency depends upon pressure drop and air flow, the size of the units is relatively large, and the most effective filters are sensitive to temperature.

Precipitators

The electrostatic precipitator is regarded as the most effectjve device for preventing the emission of dust from flue gases, and is practically standard equipment for large power stations. Unfortunately, the equipment is massive and costly to operate, so that considerations of cost limit its use for smaller boiler plants.

Scrubbers

These devices are used for particulate matter, but are better suited for vapours and gases. They may be classified as follows:

(*a*) wet washers
(*b*) gravity spray towers
(*c*) wet impingement scrubbers
(*d*) spray de-dusters
(*e*) disintegrator scrubbers
(*f*) venturi scrubbers.

Their applications in fuel processing or combustion are limited and their general application to industrial effluents is discussed elsewhere [1].

Sonic and ultrasonic agglomeration

Some attention has recently been given, particularly in the USA, to methods of agglomerating dust particles in suspension, thus facilitating their subsequent collection in a simple cyclone or wet washer. It has been standard practice for many years in the carbon-black industry to use electrostatic precipitators as pre-agglomerators. These installations are relatively expensive, and attempts have been made to produce a cheaper plant by using sonic or ultrasonic agglomerators.

The equipment consists of a generator of sonic energy, a resonance chamber in which the agglomeration takes place, and a collector for the agglomerated particles.

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[1] See page 329 (" Prevention and Control of Air Pollution by Process Changes or Equipment ", by A. H. Rose, D. G. Stephan & R. L. Stenburg).

It may be stated that sonic and ultrasonic methods of enhancing the collection efficiency of cyclones and filters have not yet proved their worth for normal industrial application.

Removal of Sulfur Dioxide

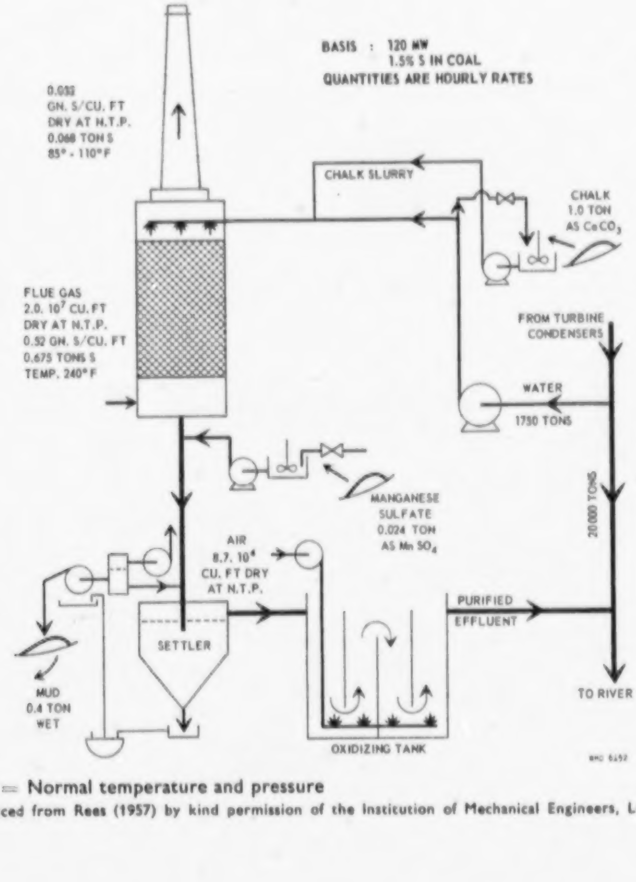
In many plants for processing sulfur-bearing ores or for manufacturing sulfuric acid, removal of sulfur oxides from the exit gases is essential. Potentially, some of these methods are applicable to the removal of sulfur dioxide from the gaseous products of combustion. But, whilst the total discharge of sulfur dioxide into the atmosphere as a result of the combustion of fuel is enormous (see Table 1), the concentration in the flue gases is comparatively low. It is the general practice, therefore, to rely upon dispersal from high chimneys to prevent the emission of sulfur dioxide becoming an undue nuisance. As far as the authors are aware, the removal of sulfur dioxide from flue gases has only been practised at a few large power stations, situated in Central London near buildings of historic importance. These processes are expensive, and the cost of applying them to smaller plants would be prohibitive. Research is being undertaken on less expensive methods of purification. Accounts of this work and of the specialized processes for removing higher concentrations of sulfur oxides in the metallurgical and chemical industries are to be found in the following papers: *Canad. chem. Process Inds*, 1942; Craxford, Poll & Walker (1952); Dean & Swain (1944); Diamond (1934); Francis (1946); Hewson et al. (1933); Johnstone (1952); Johnstone & Kleinschmidt (1938); Johnstone & Silcox (1947); Johnstone & Singh (1940); Kirkpatrick (1938); Lessing (1938); Newall (1955); Pearson, Nonhebel & Ulander (1935); Rees (1953); Rees (1957).

Rees (1957) has reviewed three processes for the removal of sulfur dioxide from the flue gases from London power stations. The first was developed by the London Power Company for Battersea. The siting of this station on the River Thames where there was a strong tidal flow led to the use of an effluent process. The river not only provides the large amount of washing water and about four-fifths of the alkalinity needed to neutralize the SO_2 in the flue gases, but also carries away to sea, in well mixed solution, the calcium sulfate produced in the process. It is perhaps the simplest flue gas washing process that could be devised, and has since been installed at Bankside power station, where it is removing 95 % of the sulfur dioxide in flue gases from the combustion of oil with a sulfur content of about 3.8 %.

In an earlier paper, Rees (1953) described the theoretical background and early development of the process. A flow diagram of its present form is given in Fig. 1, which shows the relevant quantities, on an hourly basis, for plant of 120 megawatts generating capacity burning coal with a sulfur content of 1.5 %. The effluent is oxidized by aeration in the presence of manganese in order to meet the requirements imposed by the Port of London Authority on the quality of the discharge into the river. The permissible residue of deoxygenating material in the effluent contributes only slightly to the pollution of the river. The calcium sulfate produced is capable of forming scale in certain types of heat exchanger. There is no danger of saturating the water of the Thames with calcium sulfate from Battersea and Bankside power stations, but it might be unwise to install this process at any other stations in London or for many miles downstream.

FIG. 1

FLOW DIAGRAM OF THE "BATTERSEA" EFFLUENT PROCESS


BASIS : 120 MW
1.5% S IN COAL
QUANTITIES ARE HOURLY RATES
0.032
GN. S/CU. FT
DRY AT N.T.P.
0.068 TON S
85° - 110°F
CHALK SLURRY
CHALK
1.0 TON
AS $CaCO_3$
FLUE GAS
2.0. 10^7 CU. FT
DRY AT N.T.P.
0.52 GN. S/CU. FT
0.675 TONS S
TEMP. 240°F
FROM TURBINE
CONDENSERS
WATER
1750 TONS
MANGANESE
SULFATE
0.024 TON
AS $MnSO_4$
20 000 TONS
AIR
8.7. 10^4
CU. FT DRY
AT N.T.P.
PURIFIED
EFFLUENT
SETTLER
MUD
0.4 TON
WET
TO RIVER
OXIDIZING TANK

N.T.P. = Normal temperature and pressure

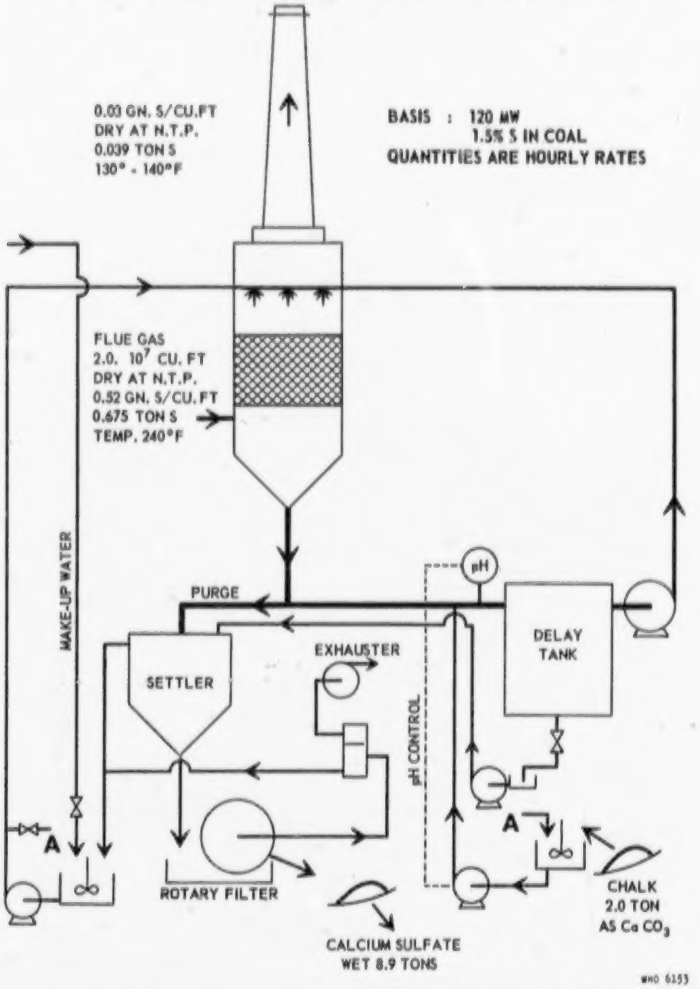
Reproduced from Rees (1957) by kind permission of the Institution of Mechanical Engineers, London.

The cost of installing, operating and maintaining the process at a modern power station adds 8-10 shillings per ton to the cost of coal burnt.

The second process described by Rees is the Howden-I.C.I. Cyclic Lime Process. Owing to the presence of oxygen in flue gases, the calcium sulfite first formed when SO_2 is neutralized by lime or chalk is oxidized to calcium sulfate. This is not very soluble, and so forms scale on scrubbers.

FIG. 2

FLOW DIAGRAM OF HOWDEN-I.C.I. CYCLIC LIME PROCESS

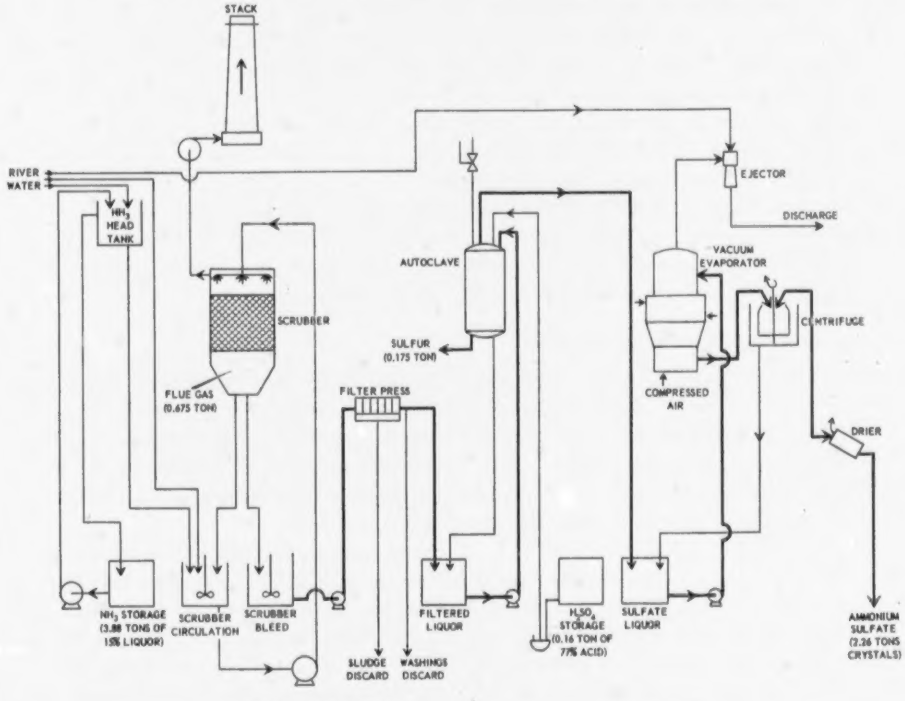

0.03 GN. S/CU.FT
DRY AT N.T.P.
0.039 TON S
130° - 140°F
BASIS : 120 MW
1.5% S IN COAL
QUANTITIES ARE HOURLY RATES
FLUE GAS
2.0. 10^7 CU. FT
DRY AT N.T.P.
0.52 GN. S/CU. FT
0.675 TON S
TEMP. 240°F
MAKE-UP WATER
PURGE
pH
DELAY TANK
SETTLER
EXHAUSTER
pH CONTROL
A
A
ROTARY FILTER
CHALK
2.0 TON
AS Ca CO3
CALCIUM SULFATE
WET 8.9 TONS
WHO 6153

Reproduced from Rees (1957) by kind permission of the Institution of Mechanical Engineers, London.

The Howden-I.C.I. process is distinguished from other cyclic lime processes by its method of overcoming this difficulty. The solution leaving the scrubbers, supersaturated with calcium sulfate, is mixed with a suspension of lime or chalk (the amount being controlled by the pH value of the solution) and sent to a delay tank, where the excess calcium sulfate crystallizes on existing crystals in suspension. The concentration of dissolved calcium sulfate is thereby reduced to saturation level, after which the slurry is pumped back to the scrubbers. The general concentration is kept roughly constant by bleeding off a fraction of the liquor and separating the suspended calcium sulfate from it. The arrangement is shown in the flow diagram in Fig. 2, the quantities being on the same basis as those in Fig. 1.

FIG. 3

FULHAM/SIMON-CARVES CYCLIC AMMONIA PROCESS FOR USE WITH AMMONIA LIQUOR FROM GAS-WORKS OR COKE-OVEN PLANT


STACK
RIVER WATER
NH_3 HEAD TANK
SCRUBBER
FLUE GAS (0.675 TON)
AUTOCLAVE
SULFUR (0.175 TON)
FILTER PRESS
EJECTOR
DISCHARGE
VACUUM EVAPORATOR
CENTRIFUGE
COMPRESSED AIR
DRIER
NH_3 STORAGE (3.88 TONS OF 15% LIQUOR)
SCRUBBER CIRCULATION
SCRUBBER BLEED
SLUDGE DISCARD
WASHINGS DISCARD
FILTERED LIQUOR
H_2SO_4 STORAGE (0.16 TON OF 77% ACID)
SULFATE LIQUOR
AMMONIUM SULFATE (2.26 TONS CRYSTALS)

FLUE GAS : 0.675 TON S
NH_3 : 3.88 TONS OF 15% LIQUOR
H_2SO_4 : 0.16 TON OF 77% ACID
SULFUR : 0.175 TON
AMMONIUM SULFATE : 2.26 TONS CRYSTALS

QUANTITIES ARE HOURLY RATES; 120 MW BASIS, 1.5% S IN COAL.

Reproduced from Rees (1957) by kind permission of the Institution of Mechanical Engineers, London.

The process removed more than 95 % of the SO_2 in the flue gases, but was costly and maintenance problems were severe. The disposal of the wet calcium sulfate sludge, amounting to considerably more than 100 tons weekly in a typical power station was also troublesome. For these reasons the process has not survived at power stations, though it is a convenient means of purifying relatively small quantities of gas.

The all-in cost of this process in a power station is estimated at 17 shillings per ton of coal burnt.

The Fulham/Simon-Carves Ammonia Process was developed to manufacture ammonium sulfate and sulfur by washing flue gases with ammonia liquor from gas-works and coke-ovens. Work on a pilot plant began in 1948 at the Fuel Research Station in order to assess the potentialities of the process. The Central Electricity Authority and Simon-Carves Limited, with the co-operation of the Fuel Research Station, have recently set up a pilot plant which handles half the gases from one of the boilers at North Wilford power station—i.e., about 1.5 million cu. ft/hr (about 42 500 m^3). The scheme is illustrated in Fig. 3. The liquor bled from the circulating system is heated under pressure with sulfuric acid to produce ammonium sulfate and sulfur. At the present time, the major field of research lies in the design of the scrubbers, which are required to remove nearly all the sulfur dioxide from the gases while retaining all but a trace of volatile ammonia within the system.

The economics of the process depend largely upon the cost and availability of ammonia liquor, upon the price that can be obtained for ammonium sulfate, and upon the cost of maintaining the plant. It is hoped that the all-in cost will be less than that of the Battersea process.

Alternatives to flue gas washing

As washed flue gas is cool and laden with CO_2 and condensed water droplets, it has a density which brings it to the ground much nearer the point of emission than if it were discharged unwashed, and therefore hot. Because of this, the public, which rarely takes notice of unwashed flue gases discharged from high chimneys, complains not infrequently about the washed flue gases from Battersea and Bankside.

As flue gas washing is also so expensive a process, much thought has been given to the possibility of extracting the sulfur from the fuel before it is burnt, or removing SO_2 from the flue gas by some method that does not cool the gas. The Central Electricity Authority has examined the possibility of the removal of pyrites from coal. As, in pulverizing, the size of the coal particles is reduced to 20-50 μ, it was hoped that the relatively dense pyrites would be split from the coal substance. Unfortunately, it has been found that it would be necessary to grind most British coals much more finely, before the pyrites particles and the coal substance could be readily separated.

The separation of sulfur from fuel oil is feasible but costly, and existing techniques are not applicable commercially to the residual oils that are used for steam generation in power stations.

Despite the amount of development work in the past, no practicable process has been found for the removal of SO_2 from flue gases without cooling the gas. The main problem of absorption by a dry reagent is the poisoning of the reagent surface by dust or sulfates and phosphates deposited from the gas.

Specific Industrial Problems

In the iron and steel industry, the position has been summarized by Thring & Sarjant (1957) as follows:

> During refining, the open-hearth furnace emits fine iron oxide fume, the emission of which is considerably increased if oxygen lancing is used. Electrostatic precipitators and Pease Anthony scrubbers have been used, but cleaning is very expensive and research on new methods is being actively pursued in the United States of America. The Bessemer also gives severe iron oxide fume and the problem of cleaning is even more difficult as the gases are not confined to a comparatively small flue. Bag filters, electrostatic precipitators and Pease Anthony scrubbers have been tried on arc furnaces and on cupolas. In the former case the comparatively small volume of gases makes the problem more soluble. Blast-furnace gas cleaning is highly developed, using systems which are economic because of the value of the clean gas.
>
> Continuous counterflow pusher-type reheating furnaces present no fume problem since the temperature is not high enough for the emission of iron oxide fume, and there is no reason to operate with smoky combustion conditions since sufficient temperature uniformity can be obtained with quite a short flame by bringing part of the fuel in at the discharge end and part further down the furnace.
>
> The furnace for reheating large ingots for forging can present a real smoke problem, not because it is necessary to have excess fuel to avoid oxidation, since the degree of surface oxidation is not great with large ingots, but because the design of the furnace necessitates delayed combustion to give even temperatures on both sides of the ingot. The solution here is clearly to change the design of the furnace.
>
> In the past, probably the greatest offender from the smoke emission standpoint has been the small hand-fired, heat-treatment furnace. A survey of heat-treatment furnaces was carried out in the Sheffield area some years ago and of 106 furnaces investigated 76 were coal fired and of these 47 were hand fired, the remainder having mechanical stokers. The heavy smoke emission coincided almost exactly with the hand-fired type of furnace... The use of mechanical firing could largely eliminate smoke formation and the change-over to town gas firing can completely eliminate it. Even with hand firing, however, it is possible to operate largely without smoke by good firing practice and overfire air jets.

Thring & Sarjant also provide an excellent outline of the problems of preventing the emission of smoke and grit from coke-ovens. Smoke arises from coke ovens and can be broadly classified as to its source under three headings: (1) during the charging and discharging operation; (2) from leaking doors; and (3) from pushing and quenching (dust as well as smoke).

(1) This can be reduced to a minimum as follows:

(*a*) by providing the telescopic connections between the charging holes in the top of the ovens and the charging car;

(*b*) by providing double collecting mains, that is, a collecting main at either end of the battery of coke ovens;

(*c*) by charging and levelling with the gas mains coupled to the oven and the use of steam ejection in the ascension pipes;

(*d*) careful attention to detail by workmen in carrying out operations correctly and quickly.

Little can be done to reduce the dust and smoke evolved during discharging. Attention to the distribution of the heating gas to the ovens is important and coal quality ought to be reasonably consistent.

(2) The latest types of self-sealing door are smokeless if the doors and frames are properly cleaned after pushing.

(3) The dust which is entrained with water vapour and steam in the quenching tower can carry some five or six hundred yards and although there is no easy method of collecting this dust, the method of quenching employed can minimize such carry-over. Instead of strong jets of water being directed on to the hot coke it has been established that a circular spray of water, which is, as it were, more gentle in its action, not only gives a better quench but also reduces the amount of dust entrained.

The foundry presents highly individual problems, which are being tackled systematically and with a high degree of success. Large quantities of dust are produced, some likely to produce silicosis. Lawrie (1957) discusses the means to be adopted for suppressing dust and fumes, and points out that the main difficulty is that many operations are conducted on a small scale, generating large numbers of particles of various sizes. He and his colleagues have developed a method for photographing normally invisible dust clouds and the technique is used to devise local exhaust ventilation systems that remove the dust through hoods.

The main danger of atmospheric pollution in the cement industry arises from the rotary kilns. A slurry of chalk and clay is fed into one end, and pulverized coal is blown in at the lower front end. To what extent the emergent dust originates from the slurry or from the coal—and therefore whether the problem lies within the terms of reference of this paper—would be difficult to determine. Burke, Murray & Johnson (1957) have reviewed methods of dust control in this industry. They discuss the application of electrostatic precipitators, which are normally used to collect dust from the rotary kilns, and compare the cost and efficiency with other methods, such as cyclone collectors.

Research on a considerable scale has been devoted to the problem of firing ceramics efficiently and smokelessly. The measures that have been taken to reduce smoke emission can be summarized as follows:

(1) Coal has been replaced by coke, oil or gas.

(2) Intermittent kilns have been replaced by continuous kilns, thus avoiding smoke emission and the waste of fuel consequent upon alternately heating and cooling the structure.

(3) Improvements have been made in the design of kilns.

(4) The standard of maintenance has been improved.

(5) The standard of operation has been improved.

(6) Mechanical stokers have been more widely used.

REFERENCES

Arthur, J. R. & Napier, D. H. (1952) *Brit. Coal Utilis. Res. Ass. Bull.*, **16,** 309

Arthur, J. R. & Smith, W. D. (1956) *Brit. Coal Utilis. Res. Ass. Bull.*, **20,** 429

Burke, E., Murray, J. J. L. & Johnson, K. R. (1957) In: *Proceedings of the conference on the mechanical engineer's contribution to clean air,* London, p. 103

Canad. chem. Process Inds, 1942, **26,** 38

Cosby, W. T. (1949) *Brit. Coal Utilis. Res. Ass. Bull.*, **13,** 225

Craxford, S. R., Poll, A. & Walker, W. J. S. (1952) *J. Inst. Fuel,* **25,** 13

Dean, R. S. & Swain, R. E. (1944) *U.S. Bur. Mines Bull.*, No. 453

Diamond, R. W. (1934) *Trans. Canad. Min. Inst. (Inst. Min. Metall.)*, **37,** 442

Dunningham, A. C. (1957) *Engng Boil. Ho. Rev.*, **72,** 40

Francis, W. (1946) *Power Wks Engr,* **41,** 17

Gunn, D. C. & Hawksley, P. G. W. (1957) In: *Proceedings of the conference on the mechanical engineer's contribution to clean air,* London, p. 77

Hawkins, J. E. & Nonhebel, G. (1955) *J. Inst. Fuel,* **28,** 530

Hewson, G. W., Pearce, S. L., Pollitt, A. & Rees, R. L. (1933) *Engineering (Lond.)*, **136,** 459

Johnstone, H. F. (1952) *Pulp Pap. (Mag.) Can.*, **53,** 105

Johnstone, H. F. & Kleinschmidt, R. V. (1938) *Trans. Amer. Inst. chem. Engrs,* **34,** 181

Johnstone, H. F. & Silcox, H. E. (1947) *Industr. Engng Chem.*, **39,** 808

Johnstone, H. F. & Singh, A. D. (1940) *Univ. Ill. Bull. Engng Expt Sta. Bull. Ser.*, No. 234

Kirkpatrick, S. D. (1938) *Chem. metall. Engng.* **45,** 483

Lawrie, W. B. (1957) In: *Proceedings of the conference on the mechanical engineer's contribution to clean air,* London, p. 128

Lessing, R. (1938) *J. Soc. chem. Ind. (Lond.)*, **57,** 373

Newall, H. E. (1955) In: Mallette, F. S., ed. *Problems and control of air pollution,* New York, p. 170

Organisation for European Economic Co-operation (1956) *Europe's growing needs of energy. How can they be met?* Paris

Pearson, J. L., Nonhebel, G. & Ulander, P. H. N. (1935) *J. Inst. elec. Engrs,* **77,** No. 463, I

Rees, R. L. (1953) *J. Inst. Fuel,* **25,** 350

Rees, R. L. (1957) In: *Proceedings of the conference on the mechanical engineer's contribution to clean air,* London, p. 34

Stairmand, C. J. & Kelsey, R. M. (1955) *Chem. & Ind.*, No. 42, 1324

Thring, M. W. & Sarjant, R. J. (1957) In: *Proceedings of the conference on the mechanical engineer's contribution to clean air,* London, p. 92

United States, President's Materials Policy Commission (1952) *Resources for freedom,* Washington, Vol. 1

ALBERT PARKER, C.B.E., D.Sc. *

AIR POLLUTION LEGISLATION: STANDARDS AND ENFORCEMENT

Introduction

Gross contamination of the air of many large towns in different parts of the world is part of the price that has so far had to be paid for the development of industry and the resultant concentration of large numbers of people in relatively small areas. It is mainly from the use of coal and its products that the air of densely populated areas in the industrialized countries is heavily polluted with smoke, grit and dust, and oxides of sulfur, though there are some areas receiving large quantities of pollutants from the manufacture of chemicals, iron and steel, cement, and other materials. Moreover, during the last fifty years there has also been an increasing amount of pollution from the use of petroleum oils for furnaces and transport.

With few exceptions, air pollution control legislation in the various countries is inadequate. A frequent disadvantage is that the first control measures were introduced many years ago, and the legislation at present in force takes no account of the enormous growth of industry, new types of air contaminant, and new sources of pollution, such as motor vehicles and heating installations. Some laws provide solely for the payment of compensation for damage caused by air pollution. In several countries, therefore, the need is becoming apparent to introduce effective legislation enforcing the adoption of adequate control measures and fixing the maximum permissible concentration of the various contaminants.

The following pages are mainly concerned with air pollution control legislation in the United Kingdom, the USA, and the USSR, but some information is also presented on the situation in certain countries of Western Europe, the source of which is a study published in 1957 by the Organisation for European Economic Co-operation (OEEC).[1] In addition, material has been extracted from the *International Digest of Health Legislation*, describing the control measures adopted in recent years in Australia, Canada, Czechoslovakia, and New Zealand.

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[1] Organisation for European Economic Co-operation, European Productivity Agency (1957) *Air and water pollution: the position in Europe and in the United States*, Paris.

United Kingdom

In the earlier years of the 19th century, when the great acceleration in industrial development began, the boilers and furnaces burning coal were not so efficient as they are today. Large quantities of smoke must have been emitted from every ton of coal burned. By 1819, the smoke nuisance was increasing to such an extent that Parliament appointed a committee to inquire if persons using engines and furnaces could erect them in a manner less prejudicial to public health and comfort. In 1843, another select committee recommended legislation to deal with nuisances from steam engines and furnaces.

Since that time, the desirability of greatly reducing the pollution of the air from the burning of coal has been stressed by many expert bodies. For example, during recent years various aspects of the problem have been considered by a Heating and Ventilation (Reconstruction) Committee of the Building Research Board of the Department of Scientific and Industrial Research, a Fuel and Power Advisory Council appointed by the Minister of Fuel and Power, and a Departmental Committee on National Fuel Policy, also appointed by the Minister of Fuel and Power. The findings of these three authorities were published in 1945,[1] 1946,[2] and 1952 [3] respectively.

In relation to air pollution by discharges from chemical and other special manufacturing processes, it should be mentioned that in the middle of the 19th century there was a public outcry as a result of the establishment of processes for making alkali from common salt. From these processes large quantities of hydrochloric acid were discharged. A Royal Commission was appointed, and following its first report, the first Alkali, etc., Works Regulation Act was passed in 1863. Later, the Act was extended on several occasions to cover a number of special processes, including many that are not related to the manufacture of alkalis.

In 1953, after the disastrous smog in December 1952, which caused the death of about 4000 people in the London area, a Committee on Air Pollution was appointed by the Government "To examine the nature, causes and effects of air pollution and the efficacy of present preventive measures; to consider what further preventive measures are practicable;

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[1] Great Britain, Department of Scientific and Industrial Research (1945) *Heating and ventilation of dwellings*, London (Post-War Building Studies No. 19).

[2] Great Britain, Ministry of Fuel and Power, Fuel and Power Advisory Council (1946) *Domestic fuel policy*, London.

[3] Great Britain, Ministry of Fuel and Power, Departmental Committee on National Fuel Policy (1952) *Report of the Committee on National Policy for the Use of Fuel and Power Resources*, London.

and to make recommendations." That Committee, which dealt with the many aspects of the subject, issued an interim report in November 1953,[1] and a final report in November, 1954.[2] Many of the recommendations of the Committee on Air Pollution have formed the basis of the Clean Air Act, 1956.

The principal legislative enactments that have been in operation in the United Kingdom during recent years, or are now in operation, are outlined in the following paragraphs.

Smoke from industrial and domestic premises

Before the passing of the Clean Air Act, 1956, the main statutory provisions in force for the control of smoke were contained in the Public Health Act, 1936, the Public Health (London) Act, 1936, and the Public Health (Scotland) Acts, 1897-1939. The authorities responsible for enforcing the Acts were: in London, the London County Council and the Metropolitan borough councils; in the rest of England and Wales, the councils of county boroughs, boroughs, and county districts; and, in Scotland, town councils and county councils.

There were also in Scotland the Smoke Nuisance (Scotland) Acts, 1857 and 1865, which applied to burghs with a population of 2000 or more, and the Burgh Police (Scotland) Acts, 1892 and 1903, administered by all town councils, except those of certain of the largest towns, who had similar powers under special local Acts.

The Public Health Act, 1936, constituted as statutory nuisances any installation for the combustion of fuel used in any manufacturing or trade process which did not so far as practicable prevent the emission of smoke; and any chimney (except a chimney of a private house) emitting smoke in such quantity as to be a nuisance. Where a notice served by a local authority requiring the abatement of a nuisance was not complied with, proceedings could be taken before a court of summary jurisdiction. The court could impose a fine of up to £50 and make an order for the abatement, or prohibition of recurrence, of the nuisance. It was, however, a defence in any proceedings for discharging smoke, other than black smoke, to show that the best practicable means for preventing the nuisance had been used.

The provisions of the Scottish and London Acts were similar. The term "smoke" was defined in the Public Health Act, 1936, as including soot, ash, grit or gritty particles. There was no definition of smoke in Scottish public general legislation. As a result, difficulties arose in attempts to deal with complaints of grit from factory chimneys.

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[1] Great Britain, Committee on Air Pollution (1953) *Interim report, presented to Parliament by the Minister of Housing and Local Government, the Secretary of State for Scotland and the Minister of Fuel and Power . . . December 1953*, London.

[2] Great Britain, Committee on Air Pollution (1955) *Report, presented to Parliament by the Minister of Housing and Local Government, the Secretary of State for Scotland and the Minister of Fuel and Power . . . November 1954*, London.

Under the Public Health Act, 1936, local authorities could make by-laws, subject to confirmation by the Minister of Housing and Local Government, regulating the emission of smoke of such colour, density, or content as may have been prescribed. Such by-laws did not apply to private houses. The by-laws usually made it an offence to discharge black smoke for a total of two minutes within a continuous period of thirty minutes. More than 200 local authorities in England had such by-laws, all of them dealing only with black smoke.

Before the passing of the Clean Air Act, 1956, there was no provision under general legislation for declaring " smokeless zones " or " smoke control areas ". About twenty local authorities, however, obtained special powers for the purpose by local Acts; the powers were used in only a few cases. These local Acts enabled the authority to define areas in which the emission of smoke was either prohibited or strictly regulated. Some local Acts also contained a " prior approval clause ", which made it an offence to install a furnace unless it could be operated, so far as practicable, without emitting smoke. The person installing the furnace, if he wished, could seek the local authority's approval to the installation in advance, and no proceedings could then be taken if the authority approved the furnace or failed to disapprove it within a specified period.

Under the Clean Air Act, 1956,[1] which is operated by local authorities, it is an offence to emit dark smoke from a chimney for longer than such periods as may be specified by the Minister of Housing and Local Government by regulations. Dark smoke, which includes soot, ash, grit and gritty particles emitted in smoke, is defined as being as dark as, or darker than, shade 2 of the Ringelmann chart. Regulations in relation to permitted periods for the emission of dark smoke, and black smoke defined as smoke as dark as shade 4 of the Ringelmann chart, have been issued.

In any proceedings for an offence under the section prohibiting the emission of dark smoke from industrial chimneys, it is a defence to prove that the contravention was due to (*a*) the lighting up of a furnace which was cold, or (*b*) some failure of a furnace or apparatus that could not be foreseen, or (*c*) the use of unsuitable fuel as suitable fuel was unobtainable. During the seven years from the passing of the Act, it is also a defence to show that the contravention was due to the nature of the building or its equipment and that it had not been practicable to alter or equip the building so that it could be used for the purpose for which it was intended without the likelihood of contravening the Act.

The Clean Air Act, 1956, also provides that no furnace, except small furnaces designed mainly for domestic purposes, shall be installed unless it can be operated, so far as practicable, continuously without emitting smoke when burning fuel of a type for which it was designed. Notice of the proposal to install the furnace must be given to the local authority, which may or may not approve the plans and specifications.

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[1] An Act to make provision for abating the pollution of the air (The Clean Air Act, 1956). Dated 5 July 1956. (*Public General Acts and Measures of 1956*, 1957, ch. 52, pp. 377-415) (published *in extenso* in *Int. Dig. Hlth Leg.*, 1958, **9**, 181).

Regulations may be made by the Minister of Housing and Local Government requiring the provision of apparatus for indicating or recording the density or darkness of smoke emitted and for making the results available to the local authority.

A local authority may, by order confirmed by the Minister, declare the whole of a district or any part of it to be a smoke control area in which only approved appliances or approved fuels or both may be used. When such an order applies to dwellings or to churches and buildings used by charities, there is provision for financial contributions from public funds towards the cost of adapting fuel-using appliances or installing new ones to meet the requirements.

Smoke from railway engines and ships

The Railway Clauses Consolidation Act, 1845, and the Regulation of Railways Act, 1868, and the corresponding Scottish Acts, provided that every railway locomotive steam engine should be constructed on the principle of consuming, and so as to consume, its own smoke; and made it an offence if any locomotive failed to consume its own smoke. No particular body was invested with the duty of enforcing these Acts. In England and Wales, any person or corporate body could complain to the courts. In Scotland, the complaint would be made to the Procurator-Fiscal, who would decide whether or not to prosecute.

Under the Clean Air Act, 1956, the provisions whereby the emission of dark smoke from industrial chimneys is prohibited apply also to railway locomotive engines. They apply, in addition, to vessels in waters not navigable by sea-going ships and in certain waters navigable by sea-going ships which are within the seaward limits of the territorial limits of the United Kingdom.

Grit and dust

The Public Health Act, 1936, constituted as a statutory nuisance any dust or effluvium caused by any trade, business, manufacture, or process, which is injurious or dangerous to the health of, or a nuisance to, the inhabitants of the neighbourhood. As with smoke emission, under the same Act it was a defence to prove that the best practicable means of prevention or alleviation of the emission had been employed.

Under the Clean Air Act, 1956, it is obligatory to use any practicable means there may be for minimizing the emission of grit and dust from a chimney serving a furnace, except for small furnaces intended mainly for domestic purposes. Further, no furnace shall be used to burn pulverized fuel, or to burn solid fuel in any other form at a rate of one ton an hour or more unless plant is provided to arrest grit and dust, which has been approved by the local authority. The Minister may direct that any application for approval under this section shall be referred to him to be dealt with by him instead of by the local authority. In addition, the Minister may, by regulations, require measurements to be made and recorded of the grit and dust emitted.

Height of chimneys

Where there are plans for the erection or extension of buildings, other than buildings to be used wholly as residences, shops, or offices, the local authority must be satisfied, under the Clean Air Act, 1956, that the height of any chimney to be constructed to carry smoke, grit, dust or gases will be sufficient to prevent, so far as is practicable, the discharges from becoming prejudicial to health or a nuisance. In reaching a decision, account must be taken of such local conditions as the levels of the neighbouring land and the position and description of nearby buildings.

Mine refuse

Under the Public Health (Coal Mines Refuse) Act, 1939, and the corresponding Scottish Act, local authorities had the power to deal with, as statutory nuisances, any deposit of refuse from a coal mine where there was cause to believe that spontaneous combustion was likely to occur. It was a defence to show that the best practicable means were being used to prevent an outbreak of fire. The Clean Air Act, 1956, provides that the owner of a mine or quarry from which coal or shale is got shall employ all practicable means for preventing combustion of the deposited refuse and for preventing or minimizing the emission of smoke or fumes.

Road traffic

The Road Traffic Acts, 1930-1947, empower the Minister of Transport to make regulations as to the construction and equipment of motor vehicles, and to the consumption of smoke and emission of visible vapour, sparks, ashes and grit. The Motor Vehicles (Construction and Use) Regulations, 1951, require every motor vehicle to be constructed, maintained and operated in such a way as to prevent avoidable emission of smoke or visible vapour. Failure to comply with these Regulations is an offence under the Road Traffic Acts. Under these Acts, action lies with the police. The Clean Air Act, 1956, does not apply to motor vehicles.

Pollutants from chemical and other special processes

Control of the emission of noxious and offensive gases from certain defined processes is exercised under the Alkali, etc., Works Regulation Act,

1906. This Act is not administered by local authorities as in the case of the Public Health Acts and the Clean Air Act, 1956, but by Alkali Inspectors, who are Government Officers appointed by the Minister of Housing and Local Government and the Secretary of State for Scotland. It includes a schedule of processes and a list of noxious and offensive gases to which it applies. The schedule and list can be modified by such Orders as seem necessary to keep pace with the developments and changes in industry. Local authorities cannot take proceedings under the Public Health Acts and the Clean Air Act in relation to the scheduled processes, without first obtaining the consent of the Minister, if action could be taken under the Alkali Act.

The Alkali Act includes the following requirements:

(1) Scheduled processes must be registered annually.

(2) A condition of registration is that the scheduled process must be provided to the satisfaction of the Chief Inspector with the "best practicable means" for preventing the escape of noxious or offensive gases to the atmosphere and for rendering such gases harmless and inoffensive.

(3) The best practicable means must thereafter be maintained in efficient working order and must be used continuously.

(4) For certain processes, upper limits are specified for the concentration of total acidity in waste gases discharged to the atmosphere.

In the schedule are included processes concerned with the manufacture or production of such materials as sulfuric acid, hydrochloric acid, nitric acid, picric acid, hydrofluoric acid, chlorine, bromine, fluorine, ammonium sulfate, and other chemical fertilizers, sulfides, bisulfites, carbon bisulfide, sulfocyanide, pyridine, tar, benzene, paraffin oil, arsenic, aluminium, lead, zinc, and cement. Electricity generating stations burning liquid or solid fuel, gas- and coke-works, and many metallurgical processes are also included in the schedule.

Electricity generating stations

Electricity generating stations are subject to control under the general Acts in relation to the control of air pollution. It is worthy of mention, however, that at two power stations, equipment is in operation for the removal of sulfur dioxide from the chimney flue gases before discharge to the atmosphere. At one of these stations, coal with only about 1 % of sulfur is being burned and, at the other, the fuel is oil containing between 3 % and 4 % of sulfur. These are special cases in which permission to build the stations included the condition that practicable methods of removing sulfur dioxide from the flue gases should be employed.

USA

In the USA, unlike the United Kingdom, there is no nation-wide or general legislation directed towards the mitigation or control of air pollution. Air pollution control is exercised under state or local powers relating to the control of nuisances and conditions affecting public health and welfare. State laws usually give authority to counties or cities. For example, California has an Act providing for the creation of " Air Pollution Control Districts " within its various counties. Such Districts are in operation in the counties of Los Angeles and Santa Clara. In the state of New Jersey there is a " Smoke Control Code " that may be adopted by county or municipal boards of health. Almost every city in the USA now has a smoke or air pollution regulation of some kind. In some instances there are inter-state compacts to improve controls, and there is an international compact between the USA and Canada to cover adjacent areas in the two countries.

Interest and action, backed by strong public opinion, have developed rapidly within the last twenty years with obvious reduction in air pollution in many areas, especially by smoke.

The American Ordinances are often more severe than the general law for the control of smoke pollution in the United Kingdom, in that they restrict the type and quality of the coals that may be used in hand-fired industrial furnaces and in domestic heating appliances. For example, the St Louis Ordinance forbids the use of solid fuel with a volatile matter content of more than 23 % or of coal of below two-inch size if it has an ash content greater than 12 % or a sulfur content greater than 2 % unless the fuels are used in furnaces of approved design which can burn them smokelessly. The emission of smoke of a density equal to or greater than Ringelmann No. 2 is prohibited, except for a maximum of six minutes in one hour in excess of Ringelmann No. 2 or nine minutes equal to Ringelmann No. 2 to allow for fires being cleaned or new fires to be started. The emission of grit and dust is limited to the equivalent of 0.45 grain per cubic foot (1 g/m^3). All plans for new fuel-burning plants and plans for the repair or reconstruction of existing plants are subject to the approval of the Smoke Commissioner. In the event of contravention of the regulations, the Smoke Commissioner has power to cause the plant to be shut down.

Other typical examples are the Ordinances of Pittsburgh and Allegheny County, which, in general, follow the same lines as those of St Louis. In Pittsburgh, however, solid fuel containing more than 20 % of volatile matter must not be used in hand-fired equipment, as compared with 23 % in St Louis. Allegheny County has a large steel industry and the Ordinance has been directed primarily towards the mitigation of pollution from industry. The limit of volatile matter for solid fuel used in hand-fired equipment is 23 %. In relation to smoke emission, there are some exceptions or relaxations for certain special processes or industries; and different maximum allowable emissions of grit are prescribed for the several industries.

The situation in Los Angeles County differs greatly from that in most other parts of the USA and even the world, in that the area of Los Angeles is subject to frequent temperature inversion, which reduces dispersion and results in the accumulation of pollutants for extended periods. Much larger quantities of petroleum oils, including motor spirit, are used in relation to the size of population than in almost any other part of the world. Little or no coal is consumed. Air pollution is caused by hydrocarbons and other products of the use of oil, and by the burning of household and garden refuse by individual householders. Recently, arrangements have been made for the centralized collection and disposal of household refuse. Legislation prohibits the discharge of smoke as dark as, or darker than, Ringelmann No. 2 for more than three minutes in one hour. Sulfur compounds may not be discharged in greater concentration than 0.2 % by volume, a restriction that is not great, since it would allow the use of oil containing 3 % of sulfur as the fuel at electric power stations. In relation to dust emission, the Ordinance provides a scale which varies with the total weight of raw material used up to a maximum discharge rate of 40 lb. (18 kg) per hour. This would be an impracticable restriction in some areas, but in Los Angeles no coal is burned, and there are few factories which emit large quantities of dust.

Steam locomotives burning coal in the USA are normally mechanically fired and much has been done to reduce smoke emission from them. Even so, they are prohibited from entering some city areas. There has been a great extension in the use of Diesel locomotives and in railway electrification.

In some areas, for example in Los Angeles, great importance is attached to the prevention of smoke emission from Diesel-engined road vehicles. Special road patrols issue warnings and institute prosecutions when necessary.

There is no doubt that the Air Pollution Control Ordinances vigorously operated with the backing of public opinion have greatly reduced pollution by smoke in many parts of the USA. At the same time, it should be recognized that the task has been facilitated by the availability of oil and natural gas at competitive prices to replace coal for many purposes, and the availability in some areas of coal containing a low percentage of volatile matter. The improvement in grit emission has not been so great, and less systematic attention has been devoted to the reduction of pollution by noxious gases from special processes than has been given by the Alkali Inspectorate in the United Kingdom.

USSR

In the USSR an Order issued in 1949 [1] stipulates that no electric power station may be constructed without the simultaneous installation of equipment for dust and ash absorption. Factories processing non-ferrous metals must likewise be equipped with devices for the absorption of dusts and gases containing compounds of sulfur, arsenic, and fluorine. Coal-tar distilleries are required to install the apparatus necessary for the absorption of hydrogen sulfide and other sulfurous gases. In iron and steel works provision must be made for the filtration of gases from blast furnaces and for the utilization of these gases as fuel, and factories using solvents must be equipped with recovery plant. Instructions issued in 1951 and amended in 1956 indicate the maximum permissible concentrations of noxious substances in urban air (see Table 1).

TABLE 1

MAXIMUM PERMISSIBLE POLLUTION LEVELS

Pollutant	Maximum permissible concentration (mg/m^3)	
	At any one time	24-hour average
Sulfur dioxide	0.50	0.15
Chlorine	0.10	0.03
Hydrogen sulfide	0.03	0.01
Carbon disulfide	0.50	0.15
Carbon dioxide	6.00	2.00
Oxides of nitrogen	0.50	0.15
Non-toxic dusts	0.50	0.15
Soot	0.15	0.05
Phosphorus pentoxide	0.15	0.05
Manganese and compounds	0.03	0.01
Fluorine compounds	0.03	0.01
Sulfuric acid	0.30	0.10
Phenol	0.30	0.10
Arsenic (non-organic compounds, with the exception of arsine)	—	0.003
Lead and compounds (with the exception of lead tetraethyl)	—	0.0007
Metallic mercury	—	0.0003

The organizational structure of air pollution control in the USSR was approved in 1950. Among the authorities responsible for supervision are the services of the State Sanitary Inspectorate, in the case of industrial undertakings subordinate to a republic, krai, oblast, okrug or municipality, and the local stations of the Sanitary and Epidemiological Department, in the case of undertakings subordinate to a rayon or city.

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[1] Order No. 431 of 14 June 1949, prescribing measures for controlling pollution of the air and for the improvement of health and hygiene conditions in urban districts. (Summarized in *Gigiena i Sanitariya*, August 1949, No. 8, p. 62) (see *Int. Dig. Hlth Leg.*, 1951, **2**, 454).

The first step in the supervisory process consists in the registration of all planned industrial activities that may cause air pollution, as well as of those factories which are existing sources of pollution. The above-mentioned authorities are also responsible for the drawing up of regulations for the prevention and control of air pollution, and for checking that the standards established during the construction or transformation of any enterprise which is a potential source of pollution have been adhered to. Newly built industrial undertakings, or factories which have undergone transformation, cannot begin operations until the necessary permit is issued by these authorities.

Supervision is exercised in the following manner. When building plans for new factories are being drawn up, representatives of ministries, central administrations, factory managers, etc., are asked to frame the measures required to prevent air pollution, and to examine and give their opinion on the plans for filtration, absorption and recovery plant, etc., and on the siting of the new enterprise. These authorities also carry out an inspection at the building stage, and are empowered to prohibit the commencement of operations if the necessary air pollution control equipment has not been installed.

The Order of 1950 makes the approval of building plans subject to the submission of particulars of the production methods envisaged, raw materials to be used, quantity and composition of liquid, solid and gaseous emissions, height of chimney stacks, etc. In its examination of projects, the supervisory authority must consider, *inter alia*, protected areas, meteorological conditions and topography, the efficacy of control apparatus, and possible changes in the quantity and quality of pollution. The sanitary authorities give their final sanction only after having ascertained that all their observations have been taken into account in the drawing up of plans. Regular sanitary inspection is carried out by the local stations of the Sanitary and Epidemiological Department.

Western Europe

Activity towards the mitigation of air pollution, in general, has not been so great in most of the countries of Western Europe as in the United Kingdom, and there has been little effective legislation, though realization of the importance of reducing air pollution is steadily increasing. The following information, describing the position in a few of the countries, is based on material published in the OEEC report, to which reference has already been made.

Belgium

There is a general regulation that prescribes the height of chimneys designed to carry smoke and flue gases containing sulfur dioxide. The prescribed height is related to the concentration of the sulfur dioxide and the temperature of the flue gases. This regulation was framed to deal with the roasting of sulfide ores and the extraction of zinc, but it applies also to other waste gases containing sulfur dioxide.

Denmark

There are no special regulations controlling air pollution. Under general legislation, it is unlawful to cause danger or nuisance to persons or damage to property by smoke, dust, obnoxious odours or noise. The Directorate of Health, which is under the Ministry of the Interior, can take action against infringement of this broad but not very precise legislation.

France

Included in regulations governing most industrial firms in France, there is a clause prohibiting the discharge into the air of dense smoke, vapour, dust and toxic or corrosive gases likely to be detrimental to public health and safety, agricultural production, public monuments and scenic beauty. There is also an Act of April 1932, which deals with the suppression of noxious industrial smoke. These regulations are not sufficiently explicit as to the degree of pollution considered to be harmful. It is only in the Department of the Seine that there are regulations in relation to furnaces burning coal or oil. They stipulate that during normal working the density of the smoke must not exceed Ringelmann No. 1 and that, with intermittent stoking, any greater smoke density must not last for more than 5 % of normal time of working the furnace. Waste combustion gases must not contain more than 1 % of carbon monoxide by volume and 2 % of sulfur dioxide. Gases issuing from chimneys must not contain more than 1.5 grams of dust per cubic metre, calculated to 0° C and 760 mm barometric pressure, and the total quantity of dust must not exceed 300 kilograms per hour. Chimneys must be of sufficient height to ensure satisfactory dilution and dispersion of gas and dust in the air. The results achieved by these various regulations have not been satisfactory, so the Government has arranged for a study of the problems of air pollution.

Germany, Federal Republic

Under the terms of a law of 21 June 1869, governing industrial establishments, the sanitary authority is empowered to take the necessary measures, at the stage of installation, to prevent any new industrial undertaking from becoming a nuisance to the population in the vicinity. If legal action is brought, the offending party is required to pay compensation for damages inflicted or to suppress the offensive agents.

It is generally agreed at the present time that such legislation is highly inadequate.

Netherlands

A local authority cannot issue a permit for the construction of plant without first consulting the regional Inspector of Works. If the Chief Inspector decides that there is a risk of undue pollution of the air, he offers a remedy which can then be enforced by the local authority. Any party concerned can appeal to the Crown against a decision by the local authority. The Crown, on the advice of the Minister of Social Affairs and Public Health, makes the final decision. The Minister is advised by experts, including officials from the Inspection of Works Department.

Portugal

No specific regulations exist to provide against undue air pollution, but the authorities concerned may suggest or enforce any action deemed necessary to prevent undue pollution. Under a Decree of August 1922, industrial chimneys must be higher by at least one metre than the highest point of the roof of any building within a radius of fifty metres from the axis of the chimney. By a Ministerial order of November, 1932, it was left to the discretion of visiting engineer inspectors to fix the height of a chimney so that fumes or smoke should not cause any inconvenience in the neighbourhood of the factory.

Sweden

There are no legal provisions in relation to air pollution. Any person suffering damage through pollution can institute a civil action against those responsible and may claim damages and the taking of measures to abate the pollution. Local health authorities have powers to prohibit the carrying on of an industrial process if the pollution caused involves obvious detriment to health. Committees have been set up to inquire into the need for more stringent regulations.

Other Countries

Australia (Victoria)

Under the Clean Air Act of 1957 [1] the emission of dark or dense smoke from industrial chimneys is prohibited, with certain exceptions, and no new industrial fire-places may be installed unless they are as far as practicable smokeless. Moreover, the emission of any other air impurities from industrial fire-places must be minimized, and new industrial fire-places must be fitted with devices to arrest such impurities. The terms "chimney", "dark smoke", "dense smoke", "fire-place" and "air impurities" are defined in detail. In addition, a Clean Air Committee is established under the Act to investigate problems of air pollution and make recommendations to the Minister as to its abatement, with special reference to power stations, locomotives, ships, aircraft and vehicles propelled by the combustion of fuel. At the executive level, the Commission of Public Health is charged with the administration of the Act, and inspectors are granted powers of entry, inquiry and inspection.

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[1] An Act (No. 6125 of 1957) to make provision for abating the pollution of the air (Clean Air Act, 1957). Dated 20 November 1957. (*Acts of Parliament, 1957*, 1958, Part II, pp. 1008-1015) (see *Int. Dig. Hlth Leg.*, 1959, **10**, 429).

Canada (Ontario)

The 1958 Air Pollution Control Act [1] of the State of Ontario gives the Minister and municipalities a variety of powers. The Minister may engage in the promotion of research on air pollution problems as well as in the framing of control measures. Municipalities are empowered to pass by-laws, subject to certain limitations, for prohibiting or regulating the emission from any source of any class or type of air contaminant. In particular, the Act defines and determines degrees of density, in accordance with which the emission of air contaminants may be prohibited or limited for certain periods of time by municipal by-laws. Municipalities may also prohibit certain combustion operations likely to produce offensive or harmful pollution. In addition, subject to certain limitations, they are empowered to regulate the installation, alteration, maintenance and operation of structures and equipment from which air contaminants may be emitted. In order to administer and enforce the policy of air pollution abatement, by-laws may be passed for the appointment of municipal officers with powers of entry, inspection, inquiry and regulation.

All the proposed municipal by-laws must be submitted to the Minister for approval.

Czechoslovakia

A decree was promulgated in 1954 [2] for the purpose of controlling atmospheric pollution, particularly that arising from the emission of dust, soot, ash, smoke, fumes and odours. The control measures to be introduced are the following: provision of suitable ventilation systems in buildings; planning and selection of the means of communication and fuel and power supplies least likely to pollute the atmosphere; suppression of atmospheric pollution around health and educational establishments, and in parks, health and recreational resorts; the siting of gardens in factory zones, residential areas and public places in order to purify the atmosphere; prevention of the emission of dust and noxious substances during building, demolition, public sanitation and industrial operations. In addition, the decree provides for the establishment of protected zones around factories and other undertakings causing atmospheric pollution.

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[1] The Air Pollution Control Act, 1958. Dated 2 March 1958. (*The Statutes of Ontario*, 1958, ch. 2, 8 pp.) (see *Int. Dig. Hlth Leg.*, 1959, **10**, 489).

[2] Decree No. 24 of the Minister of Health of 7 May 1954 on the control of atmospheric pollution. (*Sbirka zakonu republiky Československé*, 21 May 1954, No. 14, pp. 85-86) (see *Int. Dig. Hlth Leg.*, 1956, 7, 41).

The Hygiene and Epidemic Control Service is responsible for supervising the abatement of atmospheric pollution. Control is exercised through regular inspections, which include physical, chemical and biological investigation. In this connexion, also, the construction of dwellings, factories and other establishments is subject to the approval of the Hygiene and Epidemic Control Service.

New Zealand

The 1956 Health Act [1] stipulates that inspectors with powers of entry, inspection and inquiry may be appointed to investigate the situation in respect of the evolution of noxious or offensive gases in chemical works. Under this Act, the occupier of any chemical works is required to possess effective appliances for the prevention of the escape or discharge of noxious and offensive gases, or for the rendering of such gases harmless and inoffensive. Also, processes and operations in which such gases are evolved must be properly supervised.

Occupiers of fertilizer works and sulfuric acid works must, in addition to fulfilling the above requirements, ensure that acid gases of sulfur and nitrogen evolved in any of the production processes are condensed in such a way as may be prescribed by regulations.

Schedules specifying those undertakings which fall into the category " chemical works " and listing the gases termed " noxious and offensive " are attached to the text of the Act.

Conclusions

Though there have been considerable advances in dealing with problems of air pollution, with many legislative measures adopted in some countries, particularly in the United Kingdom and the USA, excessive and often unnecessary pollution is prevalent in many industrial areas. The legislative measures have not always been effective, partly because they lacked precision and legal action was beset with difficulties, and partly because ther has been insufficient public insistence on the mitigation of pollution. In the present state of knowledge, complete elimination of pollution is not practicable, but there could be much greater improvement than has so far been achieved. Such improvement will only be attained by better legislation more effectively administered, increasing knowledge based on intensified research and development work, and a more determined public opinion. As new industrial processes are developed and conditions change, modifications in legislation and methods of enforcement will be required from time to time; but it must be recognized that no one set of preventive measures will meet all needs everywhere owing to differences in the conditions in various countries and localities.

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[1] An Act (No. 65 of 1956) to consolidate and amend the law relating to public health (The Health Act 1956). Dated 25 October 1956. (*The Statutes of New Zealand, 1956*, 1957, Vol. II, pp. 959-1048) (published *in extenso* in *Int. Dig. Hlth Leg.*, 1957, 8, 643, see in particular, Part V, p. 665).

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RADIOACTIVE POLLUTION OF THE ATMOSPHERE

Radioactive pollution of the atmosphere can be defined as any increase in the natural background radiation arising out of human activities involving the use of naturally occurring or artificially produced radioactive substances. Such atmospheric pollution presents a hazard not only to those engaged in radiation work but also to the general public.

Since the discovery of artificial radioactivity and especially since the development of the atomic bomb and of techniques of harnessing nuclear energy, the chances of radioactive materials being released into the atmosphere have multiplied enormously. It is therefore of paramount importance that a thorough study should be made of the situation and adequate measures taken to ensure that radioactive pollution of the atmosphere remains below a level where it may endanger public health. For this purpose it is first necessary to examine the potential contribution to atmospheric pollution of the various sources of radioactive contamination and then to study the properties of each contaminant and its fate after release into the atmosphere. With this knowledge, it will be possible to proceed to an assessment of the harmful effects to be expected in the fields of radiobiology, radiopathology and radiotoxicology and hence to derive maximum permissible limits for atmospheric pollution. In order that these limits shall be respected, an effective monitoring system must then be introduced and suitable precautions taken to control the discharge into the atmosphere of radioactive materials.

Factors Involved in Radioactive Pollution of the Atmosphere

The first step in any study of this problem is to draw up a list of sources of radioactive pollution of the atmosphere. Then a study must be made of the quantities of radioactive materials released by each source, the types of radiation and the area of dispersion. Finally, the fate of the substances after their release into the atmosphere must be known, a complete study being made of all the changes that occur up to the final direct or indirect impact on man.

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List of sources of radioactive contaminants

The most convenient way of listing the various sources of radioactive contamination of the atmosphere is to follow the different stages in the handling of radioactive substances from their extraction and purification until their final application for peaceful or military purposes. The production of radioactive substances involves mainly heavy industries, such as the uranium and thorium mines, metallurgical factories, nuclear reactors, and chemical plants. Radionuclides find a wide variety of applications, ranging from their use as tracers or as sources of radiation for medical purposes to the production of electricity, the provision of propulsive power, and nuclear explosions. In following the various stages in the manufacture and application of radionuclides, it is of interest to consider the object of each procedure, the radioactive wastes which are obtained, and the continuous or occasional atmospheric pollution which may result.

Production of nuclear fuel

The production of nuclear fuel for subsequent use in reactors or for nuclear explosions involves the following successive stages: mining of the crude uranium or thorium ore; washing and concentration of the ore in processing plants adjacent to the mines so that it can be transported economically; production of ingots of refined uranium or thorium at the metallurgical works; and chemical separation of the different isotopes of uranium and thorium. A common feature of all these operations is that they involve only naturally occurring radioactive elements belonging to the uranium and thorium families.

During these production processes, considerable quantities of wastes accumulate: in the mines and adjacent processing plants, these take the form of heaps of solid deads containing a very small percentage of ore; in the metallurgical works, the residues may be solids or sludges and contain all the daughter products of uranium and thorium, in particular all the radium. The production process may thus be split into two phases, one preceding and the other following the separation of uranium and thorium from their daughter products. Each of these phases will make a different contribution to atmospheric pollution.

The phase preceding the separation of uranium and thorium from their daughter products comprises the mining of the ore and the preliminary processing carried out in the plant adjacent to the mines (washing, concentration) and in the metallurgical works (crushing, milling). Atmospheric pollution may occur in the course of these operations as the result of the release of radioactive gases (radon, thoron), the adsorption of solid daughter products of radon and thoron on particles suspended in the atmosphere, and the formation of dusts from uranium or thorium ores.

The phase following the separation of uranium and thorium from their daughter products comprises the purification of metallic uranium and thorium, after passage through various salts or other compounds, and the separation of uranium and thorium isotopes, which is usually effected by the gaseous diffusion method. It is characteristic of this phase that atmospheric pollution by the radioactive gases radon and thoron cannot occur except when they are evolved by solid wastes or sludges stored near the metallurgical works. Pollution resulting from these operations is usually due either to dusts of uranium and thorium oxides or to gaseous compounds, especially uranium fluoride.

Operation of nuclear reactors

The nuclear fuels thus obtained are introduced into reactors, where three noteworthy processes take place:

(1) Fission processes: the primary fuel is partially transformed into secondary fuel (e. g., uranium gives rise to plutonium) with the simultaneous appearance of fission products.

(2) Activation processes: stable substances introduced into the channels of the reactor are activated by the neutron flux and partially transformed into radionuclides.

(3) Thermal processes: during the fission and activation processes heat is generated necessitating the use of coolant systems; by means of heat exchangers, the heat can be converted into other forms of energy, particularly electricity.

Radioactive wastes formed during these processes are of two types: fission products, which remain incorporated in the primary and secondary fuels; and extraneous activation products, found mainly in the coolant. Both the fuel elements and the coolants are thus potential sources of radioactive atmospheric pollution. This may come about through the release into the atmosphere of radioactive gases, such as xenon and krypton, produced by fission; through the induced activity of atmospheric argon; through the formation of radioactive aerosols containing primary or secondary fuels (uranium, thorium, plutonium); through the release of fission products (^{90}Sr, ^{144}Ce, ^{140}Ba, ^{95}Zr, etc.); or through induced activity of various kinds. Thus, although atmospheric pollution with naturally occurring radioactive elements may result from the operation of nuclear reactors, the main risk is from radionuclides produced artificially by fission or activation.

Chemical and metallurgical processes

The aims of chemical and metallurgical processing of the products recovered from nuclear reactors are (*a*) to separate the primary fuel, the secondary fuel, and the products of fission and activation; (*b*) to recover the primary fuel (uranium and thorium) and convert it into usable metal; (*c*) to extract the secondary fuel (e. g., plutonium) and prepare metallic products or chemical compounds from it; and (*d*) to convert the fission and activation products into radionuclides for use in industry, research, and medicine.

The radioactive wastes resulting from these processes are very important since they comprise all the unwanted excess fission and activation products as well as the activation products derived from impurities. As these wastes are produced in large quantities and have a high activity, they pose an extremely difficult public health problem, especially as regards their treatment and storage.

Fortunately, the risk of atmospheric pollution is relatively small, since most of these operations are carried out in the liquid phase. The only hazard, therefore, is from volatile substances or liquid aerosols, except for the dusts produced during the machining and metallurgical processing of the metals (uranium, thorium, plutonium, etc.). At all events, pollution is due almost entirely to artificially produced radionuclides, mainly in the form of fine particles suspended in the atmosphere. The radionuclides involved are generally alpha- or beta-gamma-emitters with a long half-life.

Peaceful uses of radionuclides

The applications of radionuclides in industry, agriculture, medicine and scientific research are becoming so extensive that only the principles on which they are based can be indicated here.

One application is the use of sealed sources either for gammagraphy or for massive external irradiation (sterilization). Secondly, radionuclides may be used in the form of sources, either as tracers in industry, biology or agriculture, or for internal irradiation in medicine. Radioactive wastes result only from the second type of application. They may be either the unused remains of the radionuclides employed, or transformation or excretion products, and the quantities involved are small. The wastes may be in the form of liquids, solids, or sludges, but are rarely gaseous. Atmospheric pollution is generally of little significance, except where dispersal over a wide area is liable to occur, such as in agriculture, iron and steel industries, and oceanography. The pollutants are usually in the form of soluble or insoluble compounds suspended as fine particles in the air, and are mostly beta-gamma-emitters having a short or medium half-life.

A possible future development that must be borne in mind is the use of radionuclides as a source of propulsive power. This will raise similar problems to those presented by a nuclear reactor, but with the additional complication that the source of contamination will be mobile.

Nuclear tests

There is one other source of radioactive pollution of the atmosphere—namely, tests of nuclear explosives. These are of two types:

(1) those based on fission processes, employing ^{235}U or ^{239}Pu;

(2) those based on fusion reactions, employing light nuclei (hydrogen or lithium).

The explosion takes the form of a non-moderated chain reaction, in the course of which a very large neutron flux appears, capable of rendering the surrounding matter radioactive. The radioactive products released in such an explosion thus comprise: unused explosive (^{235}U and ^{239}Pu), fission products formed from these explosives (^{90}Sr, ^{137}Cs, ^{131}I, etc.), and activation products formed by neutron bombardment of elements present in soil or water (^{45}Ca, ^{24}Na).

The force of the explosion and the very considerable rise in temperature that accompanies it convert these radioactive substances into gases or else eject them high into the atmosphere in the form of more or less fine particles, the height attained being greater for fusion bombs than for fission bombs. The immediate result is thus a primary pollution of the atmosphere at the site of the explosion. This is followed, however, by a secondary pollution due to radioactive fall-out. The distance covered by the particles of radioactive material will vary with the height to which they are ejected and with their size. They will eventually settle out or be carried down by rain and become dispersed over the surface of the ground. In this way, pollution is produced at points more or less remote from the site of the explosion and after varying periods of time, depending on the type and power of the bomb. After a certain time has elapsed, the principal contaminants remaining are ^{90}Sr and ^{137}Cs.

Quantitative and qualitative aspects of pollution

The importance of radioactive pollution of the atmosphere must be considered in relation to the resulting hazard. This depends not only on the actual level of pollution but also on the toxicities of the radioactive substances. In addition, it is necessary to take into account the area of dispersal and the frequency with which the release of radioactive materials occurs.

Level of radioactive pollution

From the quantitative aspect, it is useful to consider both the total quantity of radioactive material released into the atmosphere, expressed in curies, and the concentration per unit volume of air, expressed, for example, in curies per cubic metre. It is evident that quite large quantities of radioactive substances may be present in the atmosphere without constituting a hazard, provided that the specific activity per unit volume of air is sufficiently low. This is, in fact, the principle underlying the method of dispersal for the control of atmospheric pollution. Thus, the relatively large amounts of radioactive substances discharged from the tall chimneys of nuclear reactor installations result in only a low atmospheric concentration of radioactive substances in the neighbourhood. On the other hand, in a mine gallery, where the volume of air is very limited, even small quantities of radioactive materials are sufficient to produce a dangerous concentration.

Toxicities of radioactive pollutants

Neither the total quantity of radioactive material released nor the concentration per unit volume of air provides in itself a sufficient indication of the dangers involved since radionuclides vary widely in toxicity. For this reason, atmospheric pollution by the rare gases (e. g., ^{41}A) which are beta-gamma-emitters does not constitute a serious hazard, even when the activity is considerable, whereas pollution by alpha-emitters (daughter products of radium, or plutonium) may be dangerous in quite low concentrations. This explains why atmospheric pollution is much less dangerous in laboratories handling fission products than in mine workings and factories engaged in the refining of uranium and plutonium. The danger is still less in laboratories employing radionuclides with a short half-life, such as ^{32}P or ^{131}I.

Area of pollution

Even with a knowledge of the concentrations of radioactive substances in the atmosphere and their toxicities, it is still not possible to form a final assessment of the hazards resulting from atmospheric pollution. A factor of particular importance is the area affected by the pollution. Obviously, the larger the number of persons exposed to contamination, the greater is the risk to public health. As will be seen later, in fixing maximum permissible levels for radioactive pollution of the atmosphere a distinction is made between workers subject to occupational exposure and the general public, because certain serious hazards, such as the induction of genetic mutations, attain an overriding importance as soon as large populations are involved.

Areas of atmospheric pollution may be graded as follows:

Local pollution. This implies that pollution is confined to the area immediately surrounding the site of emission of the radioactive substances. Examples are mine galleries, metallurgical laboratories producing uranium or plutonium, and laboratories employing radionuclides in the gaseous phase. Only the workers on the spot are liable to be exposed to contamination, so that the numbers involved are small. This is one of the reasons why work may be permitted under such conditions, provided that suitable precautions are taken.

Regional pollution. This term is applied to radioactive pollution extending beyond the immediate surroundings of the industrial establishment. The area affected usually has a radius of several kilometres. Examples are atomic power stations and establishments for the production of radionuclides. The height of the factory chimneys is so calculated that the dispersal of radioactivity is sufficient to ensure a final concentration below the danger level. In these cases, both the employees working at the nuclear installation and the population of the surrounding areas are liable to be exposed to atmospheric pollution. In general, the number of persons affected does not amount to more than a few thousand, while secondary contamination of the soil, vegetation, and animals occurs only over a restricted area. Nevertheless, the possibilities of regional pollution should be borne in mind when selecting nuclear reactor sites.

General pollution. The peaceful uses of atomic energy can only lead to general atmospheric pollution through the additive effect of a large number of sources of regional pollution, such as might occur if there were a vast increase in the exploitation of nuclear energy. On the other hand, the atmospheric pollution brought about by nuclear tests is not only very great in the areas adjacent to the site of the explosion but is detectable all over the world. Radioactive fall-out, consisting mainly of ^{90}Sr and ^{137}Cs, occurs over a wide area and a long period, the radioactive deposits finding their way into the food chain. Thus the danger lies less in the degree of activity, which is relatively small, than in the extent of the polluted area.

Frequency and duration of pollution

It is rare for atmospheric pollution to be a constant or regular phenomenon. In the majority of cases, considerable fluctuations occur. It is useful to distinguish between continuous, accidental, and occasional pollution.

Continuous pollution. Most of the operations already referred to, whether they take place in uranium mines, nuclear reactors, chemical or metallurgical works, lead to continuous pollution of the atmosphere. In certain cases, the pollution is remarkably constant, e. g., the operation of nuclear reactors having closed-cycle coolant systems is accompanied by small losses of coolant which lead to steady pollution of the atmosphere. On the other hand, the pollution arising from certain other types of radiation work, although continuous, may be subject to large fluctuations. In uranium or thorium mines, for example, the degree of contamination varies considerably, depending upon whether a new gallery is being drilled, blasting is in progress, or the ore merely being carted away. Under such conditions, it is often difficult to make a correct assessment of the average radioactive pollution over a given period of time.

Accidental pollution. The public health worker must concern himself not only with the continuous pollution of the atmosphere arising in the normal course of radiation work but also with the possibility of accidental pollution caused by the faulty operation of the installations concerned. Although the possibility of such accidents is practically excluded for uranium mines, it must be anticipated in the case of chemical and metallurgical works and still more so in the case of nuclear reactors. In fact, fracture of the closed-cycle coolant system in a nuclear reactor may cause a sudden, massive pollution of the atmosphere with activation products formed in the coolant. Only relatively low activities are involved, however, and the radionuclides released are not usually very toxic. More serious accidents may occur if the nuclear fuel takes fire. This leads to pollution by very toxic radionuclides, such as plutonium and fission products. Moreover, the quantities of radioactive materials released may be considerable and capable of polluting the atmosphere over a wide area around the installation. This is a possibility which has to be taken into account when selecting sites for certain nuclear reactors, and it also creates a problem that will merit attention in the future if small mobile reactors are developed which can be used to provide nuclear propulsion.

Occasional pollution. This may arise as the result of isolated experiments or tests, e. g., in the use of radioactive tracers in the iron and steel industry, or in agriculture. The hazards of such pollution must be considered in relation to the frequency with which the releases of radioactive substances occur. This also applies to nuclear tests, but in this connexion it must be pointed out that the pollution is "occasional" only close to the site of the explosion. In spite of the fact that the nuclear explosions take place only at intervals, the general pollution of the atmosphere takes on a permanent character owing to the long-term radioactive fall-out, although there may be fluctuations in intensity depending on the meteorological conditions and especially on rainfall.

Fate of the radioactive contaminants present in the atmosphere

It is important to study the fate of the radioactive contaminants after their release into the atmosphere, since on this will depend the final consequences of atmospheric pollution for man and his environment. The course of events is governed partly by intrinsic factors related to the nature of the contaminants and partly by extrinsic factors dependent upon the state of the environment.

Intrinsic factors

The properties of radioactive contaminants may be classified in two main groups—physicochemical properties and specific radioactive properties.

Physicochemical properties. Every radioactive contaminant comprises a vehicle of the radioactivity which has certain characteristic physical and chemical properties.

This vehicle may be present in a variety of physical forms and may be composed of single atoms in the case of the rare gases, of molecules of more or less complex chemical compounds, or of aggregates of molecules. Two physical forms are particularly interesting—namely, the gaseous and the particulate. Radioactive gases and vapours diffuse very readily and behave very much like air in the subsequent course of events, particularly after inhalation by man. The particles in suspension in the air may be present in the form of liquid or solid aerosols. It is clear that any physical events affecting these aerosols, such as diffusion, sedimentation, absorption, or adsorption, will be conditioned by certain properties related to the dispersed state, in particular the particle size.

The chemical nature of a radioactive compound likewise influences the fate of the radioactive element of which it is the more or less temporary vehicle. During subsequent chemical reactions, the chemical affinities of the radionuclide itself become the predominant factor in determining its ultimate fate. All too often, it is only these chemical affinities which receive consideration, whereas, in fact, the properties of the material vehicle of the radionuclide—especially its solubility—are at least equally important. The fate of ^{32}P, for example, may be totally different, depending upon whether it is present in the form of a colloidal phosphate, an insoluble phosphate, or a soluble phosphate.

Radioactive properties. These influence the fate of the radionuclide only at a secondary stage. The main characteristics are decay of the radionuclide and the accompanying emission of radiations.

Broadly speaking, the decay of radioactive elements is governed by an exponential law. Each radionuclide has a characteristic decay constant, which is usually expressed in terms of the half-life, i. e., the time necessary for half the atoms to decay. This period may range from a few seconds to hundreds of thousands of years. It follows that, in the case of a radionuclide with a short half-life, the radioactivity rapidly dies out by itself, provided that no fresh pollution occurs; whereas, if the pollutant has a long half-life, radioactive pollution may remain practically constant, other things being equal. Radioactive elements can emit three types of radiation: alpha rays carrying a positive charge, beta rays carrying a negative charge, and electromagnetic gamma rays. The energy of this radiation may vary from a very low value to several million electron-volts. The effect on living organisms depends largely on the type of radiation emitted, especially on its penetrating power, which is weak for alpha rays, medium for beta rays, and strong for gamma rays.

Environmental factors

The fate of radioactive contaminants is also influenced by environmental conditions. These may be placed in two groups, namely—conditions related specifically to the atmosphere, and conditions of a broad ecological nature.

Atmospheric conditions. In order to determine what changes will take place in atmospheric pollution a knowledge of the atmospheric conditions is essential. These conditions will affect the rate of diffusion of the radioactive contaminants, the turbulence to which they are subject, and the rate of sedimentation and deposition. In particular, the area over which the pollution will spread and the degree of uniformity attained in the concentration of radioactivity will depend on atmospheric conditions. It is convenient to consider these conditions from the point of view of (1) confined spaces, such as working premises, and (2) the open air.

(1) *Confined spaces.* The conditions of stagnation, renewal, or turbulence of air in a confined space must be known before it is possible to assess what changes are likely to occur in radioactive pollution. There are great differences between the relatively calm air of a chemical laboratory, the frequently renewed air inside the buildings of metallurgical works, and the turbulent air in the galleries of a uranium mine. In the chemical laboratory the conditions favour the settling out of the contaminant on the floor and walls, whereas in the turbulent air conditions of the mine gallery settling is prevented and the particles are maintained in suspension.

(2) *Open air.* In the open air, the decisive factors are the meteorological conditions, particularly the speed and direction of the wind, the humidity, and temperature gradients. It is useful to consider separately the effects of these conditions on regional pollution and on general pollution.

The changes that take place in the radioactive pollution of the atmosphere in the neighbourhood of a nuclear reactor are governed by regional, i. e., micro-climatic, conditions. In particular, it is necessary to have an accurate knowledge of the prevailing winds and of the frequency of temperature inversions. In selecting sites for the erection of atomic installations, it is on the basis of these meteorological factors that an assessment should be made of the hazard to the local population from permanent or accidental radioactive pollution.

On the other hand, in the study of the general pollution of the atmosphere caused by nuclear tests, the climatic conditions must be considered on a global scale. The dispersal of the radioactive products in the upper layers of the stratosphere is tantamount to general diffusion; passage from the stratosphere to the lower layers of the atmosphere takes place according to laws which are still imperfectly known. It is only in the lower layers, where the atmospheric disturbances are best known, that it is possible to study the fate of the radioactive contaminants, particularly in relation to rainfall.

Ecological conditions. Radioactive pollution of the atmosphere is inevitably followed by an exchange of radioactive contaminants with the biological environment. This exchange may take place either directly or indirectly.

Many living organisms can become directly contaminated on inhaling air polluted by radioactive substances. This is true of all air-breathing animals, particularly man. Contamination occurring in this way is characterized by the fact that the secondary concentration of radioactive substances inside the body depends only on the selective power of fixation of organs for which the radionuclides show a special affinity. Apart from exceptional cases, the concentrations attained are relatively low.

On the other hand, in the case of indirect contamination, the contaminating radionuclide may follow an extremely complicated path, passing right down the food chain. Atmospheric pollution is naturally followed by deposition of the contaminants on the ground or on surface water. The radioactive substances penetrate more or less deeply into the soil which may serve, at least temporarily, to filter out the radioactivity. On the other hand, water, if secondarily contaminated, generally acts as a medium for the spread of radioactivity.

The contamination is passed on to vegetation and animals, which draw their nourishment from the soil and water. The fate of the contaminant will depend on the biological cycle peculiar to each organism as well as on successive exchanges between the vegetable and animal species. During these exchanges, secondary concentrations—often high ones—will be produced in certain organisms. Thus the specific radioactivity of seaweed may easily be 1000 times that of the water surrounding it, and in the case of plankton, the concentration factor may reach 5000. It is evident that in aquatic animals feeding on these organisms, the concentrations may become still higher. Generally speaking, land plants tend to concentrate radioactivity more in the leaves and stems than in the seeds. This is a factor which is to the disadvantage of herbivorous species as compared with those which live on grain or fruit.

In the case of man, the varied diet which he enjoys multiplies the sources of contamination. At present, milk must be considered one of the principal vehicles of indirect contamination in European countries. This explains why the bones of children, who have a predominantly milk diet, exhibit a higher ^{90}Sr content than the bones of adults.

Man is at the end of the chain of reactions and interactions undergone by radioactive contaminants. There are two main routes by which the resultant final irradiation of the body is brought about:

(1) direct, arising either from exposure to the radiations emitted by radioactive gases or suspended dust, or from radioactive contamination of the skin or respiratory tract;

(2) indirect, arising from internal contamination through the digestive tract, after the passage of the radioactive contaminants through the food chain.

Direct exposure or contamination of the first type occurs only where there is an occupational hazard or in the immediate neighbourhood of nuclear reactors, whereas indirect contamination of the second type involves whole populations.

Effects of Radioactive Pollution of the Atmosphere

In a consideration of the effects that radioactive pollution of the atmosphere may be expected to produce on man and his environment, the effects on man are obviously of the greater interest. The first part of this section will therefore be devoted to radiotoxicology, and the remainder to the fundamental question of establishing maximum permissible doses of radiation.

Radiotoxicology

Basic principles of radiobiology

To facilitate understanding of the effects of radioactive pollution of the atmosphere, it is necessary to recall briefly the basic principles governing the biological action of ionizing radiations.

Every radiobiological effect is due to the transfer of energy from radiation to living matter. The most important factors in establishing cause-and-effect relationships between exposure to radiation and its sequelae are as follows:

Quantity of energy set free. It is clear that the biological effect obtained depends on the amount of energy set free in the tissues. However, the relationship is not necessarily a linear one. Furthermore, this single quantitative aspect is not sufficient to explain all the observed facts.

Topographical distribution. The topographical distribution of the energy set free in the body is an essential factor, since the same amount of energy may have completely different effects according to whether it is concentrated at a single point or dispersed throughout the whole body.

Time factor. In certain cases, but not in all, the period of time over which the energy is transferred to the body plays a decisive role, the effects produced by a given amount of energy being smaller the longer the period of time involved.

Type of radiation. For the same amount of energy released under the same conditions of space and time, different types of radiation may have very dissimilar biological effects. Generally speaking, the relative biological effectiveness of any given type of radiation is a function of the ionization density of its constituent particles (e. g., alpha rays are more toxic than beta rays).

Nature of the tissues. Other things being equal, the same amount of energy set free in two different types of body-tissue may bring about dissimilar effects.

The tissues vary in their sensitivity to radiation, the most sensitive being called critical tissues or organs (blood-forming organs, digestive mucosa, skin, gonads).

It follows from these considerations that the concept of "exposure dose" is a complex one and must be related to a certain quantity of energy set free in a given volume during a given time, the nature of the incident radiation and the tissue irradiated being also specified.

The unit employed to express the dose absorbed is the "rad", corresponding to the liberation of 100 ergs per gram of substance irradiated. Taking into account the nature of the incident radiation the dose absorbed may also be expressed in "rem"; this value is obtained by multiplying the dose expressed in "rad" by the relative biological effectiveness of the radiation concerned.

Basic principles of radiopathology

Radiopathology is the study of the adverse effects of ionizing radiations on health. These pathological effects are subject to the same laws already described for general biological effects of radiation. They exhibit certain general characteristics which may be usefully considered here.

Polymorphism. Exposure of the body to radiation may give rise to an immense variety of biological effects: functional disturbances, such as erythema or glandular hyposecretion; morphological changes, such as fibrosis of the connective tissues or anaemia; cancers, such as epithelioma, sarcoma or leukaemia; shortening of the life-span and premature aging; and genetic defects. These disturbances are generally divided into somatic effects, which appear in the irradiated individual himself, and genetic effects, which appear in his descendants.

Latency of the manifestation. One very general characteristic of the effects mentioned above is that there is a more or less long latent period before they make their appearance. In certain cases of acute exposure, this latent period may be no more than a few days or a few hours, whereas in subacute cases it is of the order of a few weeks. As regards carcinogenic effects, several years or decades may elapse between the time of exposure and the appearance of cancer. This delayed effect is even more marked in the field of genetics; although defects involving dominant genes may appear even in the first generation, those involving recessive genes are spread over scores of generations. It is customary to distinguish between early effects (benign functional or morphological changes) and late effects (shortening of the life-span, production of cancers and genetic mutations).

Probability of appearance. The probability of the appearance of pathological changes after a given dose of radiation is dependent on dose-effect relationships. In the case of functional disturbances and benign lesions there is almost always a threshold dose below which no symptoms appear. On the other hand, even very weak doses may produce shortening of the life-span, and a significant increase in leukaemia and genetic mutations, the frequency of these effects increasing with the dose received. Indeed it has now been established that, at least within certain limits, there is a linear relationship between the dose absorbed and the effect obtained.

Reversibility. In the vast majority of functional disturbances and benign morphological changes, the cells and tissues are capable of partial or complete recovery; in particular, in the case of chronic exposure leading only to mild functional disturbances, it is possible to envisage complete recovery if the dose rate is sufficiently low. On the other hand, as a rule, only partial recovery is possible after morphological changes, while cancerous changes and genetic mutations are invariably irreversible.

Transmissibility. Transmissibility is the essential characteristic of the defects associated with gene mutations. However, transmissibility may also be found as a cellular phenomenon, especially in the case of cancer. On the other hand, functional disorders and benign morphological lesions are not transmissible.

These general characteristics underline the gravity of the pathological effects of radiation. The most serious diseases are those which are irreversible and transmissible, the absence of a threshold dose and the prolonged latent period considerably increasing their disquieting nature. Functional disturbances and morphological changes, such as skin burns and severe anaemia are often much more striking to the layman than, for example, leukaemia or genetic mutations, but it is the latter that present the most serious public health hazard.

Types of exposure

There are two major types of irradiation of the body; exposure to radiation from distant sources, and contamination by radioactive substances which come into contact with the skin or find their way inside the body.

Both types of exposure may result from radioactive pollution of the atmosphere, but in very different degrees.

Exposure to external radiation can occur only in the case of pollution by radionuclides emitting gamma rays, each radioactive atom in suspension in the air constituting a source of external exposure. Examples of such external sources are solid decay products of radon in suspension in the air and radioactive gases, such as ^{41}A.

The most serious hazard, however, is from radioactive contamination. This may be either external or internal.

External contamination arises when radioactive particles suspended in the atmosphere are deposited on the surface of the skin. This may result in cutaneous irradiation; in whole-body irradiation in the case of gamma-emitters; or in internal contamination via the respiratory or digestive tract when the particles do not adhere very firmly to the surface of the skin.

The respiratory route is the direct one and by far the commonest and most important. The digestive route is indirect, and is the one followed after secondary contamination of foodstuffs or sometimes after cutaneous or respiratory contamination. Internal contamination through the skin is exceptional and usually follows injury.

Generally speaking, persons engaged in radiation work will be subject to a different type of exposure from the population at large. They will be particularly liable to skin and respiratory contamination, whereas the main hazards for the general population are from direct exposure to radioactive substances in the atmosphere and from indirect contamination via the digestive tract. These hazards, however, remain considerably lower than those to which radiation workers are exposed.

Metabolism of radioactive substances

Radiotoxicology is dominated by the metabolic fate of the contaminants within the body, apart from the relatively rare cases of whole-body exposure to an external source (environmental gamma-radiation).

In the case of radioactive contamination, the effects depend on the routes of entry of the radionuclides and on their distribution and fixation within the body, as well as on the mode of elimination. It has already been shown that, in addition to ordinary physical and chemical properties, the radionuclides have special properties depending on the instability of the nuclei—in particular they undergo decay and emit radiation of a certain type and energy. It is clear that the metabolic fate of the radionuclides will depend exclusively on their physicochemical properties, whereas the pathological consequences will depend on their radioactive properties.

In assessing the dose absorbed by the body, it is necessary to bear in mind not only the amount of radionuclide introduced, but also the distribution of the radiation over the body and the duration of exposure, as well as the nature of the rays emitted and the sensitivity of the tissues involved.

As concerns the space-time distribution of the radiation, the phenomena are complex, involving the whole range of properties of the radionuclide concerned.

Thus, the primary topographical distribution is related to the physical form and chemical affinity of the material vehicle of the radioelement—for example, insoluble aerosols settle in the respiratory mucosa whereas gases or soluble products spread throughout the body and become fixed to organs for which they have a preferential affinity.

Superimposed on this primary distribution of the radioactive sources is a secondary distribution, which is dependent on the nature and energy of the particles emitted—for example, very short-range irradiation of a few microns in the case of alpha-emitters, local irradiation of a few millimetres in the case of beta-emitters, regional or general irradiation in the case of gamma-emitters. Similarly, the duration of exposure to the radiation is dependent primarily on the physicochemical properties of the radionuclide, which may be evaluated, as a first approximation, in terms of "biological half-life". This is the time necessary for half the radionuclide to be eliminated from some given organ or from the whole body. By way of example, those elements having a rapid turnover, such as phosphorus and sodium, may be contrasted with elements remaining in the body for a long time, such as strontium and plutonium. Secondly, the duration of exposure depends on the rate of decay of the radionuclide, which is normally characterized by its half-life, i. e., the time necessary for half the radioactive atoms to decay. Here again, radionuclides with a short half-life, such as ^{24}Na and ^{32}P, may be contrasted with those having a long half-life, such as ^{90}Sr and ^{239}Pu.

The interaction between physicochemical and radioactive properties finally results in a complex topographical distribution of the radioactivity over the body and in a duration of exposure which is expressed as the "effective half-life"—the resultant of the biological and radioactive half-lives.

Toxicological classification

From the preceding considerations it follows that, for toxicological purposes, the radionuclides can be classified in accordance with the extent and seriousness of the injuries which they cause. In a perfect classification, the comparative toxicities of the radionuclides would be ranked in relation to the physicochemical form in which they occur. However, work on this question has only just begun, and toxicological classification is in most cases based only on the radionuclides in a soluble, readily assimilable and diffusible form.

The relative toxicities of the radionuclides depend both on their physicochemical and their radioactive properties, i. e., on their chemical affinities, fundamental metabolic effects, biological half-life, radioactive half-life, and the type and energy of the radiation emitted. It may be said that, in general, the most dangerous radionuclides are those that have, in addition to a selective affinity for particularly radio-sensitive tissues, long biological and radioactive half-lives, particularly those emitting radiation with a high ionization density. Thus, ^{24}Na and ^{32}P have relatively low toxicities because they distribute themselves in a relatively uniform manner throughout the body, their biological and radioactive half-lives are short, and they are beta-emitters. On the other hand, ^{90}Sr and ^{239}Pu are very toxic, because they have a selective affinity for the bones, exposing the particularly vulnerable bone marrow to radiation, while their biological and radioactive half-lives are long.

Furthermore, plutonium, being an alpha-emitter, is even more toxic than strontium, which is a beta-emitter.

Maximum permissible doses

The first task facing public health authorities is the establishment of the maximum permissible levels of radioactive pollution of the atmosphere. To facilitate understanding of the problem, the general question of establishing maximum permissible doses of radiation will be considered first; the fixing of maximum permissible levels for radioactive pollution of the atmosphere can then be dealt with merely as a special case of exposure to radiation.

Fundamental considerations

The first question to be decided before maximum permissible doses can be fixed is whether even small amounts of radioactivity can be considered harmless, tolerable or permissible. The answer to this question will depend upon whether the somatic or genetic effects of the radiation are being considered.

Somatic effects. From this point of view, radiation may be accepted as tolerable if no adverse effects become apparent during the lifetime of the individual exposed to it. In the case of reversible and non-transmissible effects appearing only above a certain threshold dose, the expectation of natural recovery makes it possible to envisage a maximum permissible level of radiation corresponding to the threshold dose. On the other hand, in the case of irreversible effects, particularly those for which there is no threshold, there can be no permissible level of radiation and one is forced to apply the same arguments as developed below for genetic effects.

Genetic effects. In view of the irreversibility and transmissibility of these changes and, especially, since there is a linear relationship between dose and effect, radiation nearly always leads to genetic damage. Nevertheless, it is possible to consider the genetic effects as tolerable if, on the one hand, the rate of spontaneous mutations is increased only to a small extent and, on the other, only a small fraction of the population is exposed to the radiation.

Methods of establishing maximum permissible levels

Thus, maximum permissible levels of radiation may be defined in two ways: (*a*) by fixing the maximum permissible threshold dose corresponding to the appearance of benign somatic effects; and (*b*) in the case of cancer or genetic mutations, by comparing the increase in the rate of appearance with the spontaneous rate or with other risks of modern life.

The first method is the one generally used by the International Commission on Radiological Protection in establishing maximum permissible levels for occupational exposure, i. e., levels of radioactivity that are considered incapable of causing any appreciable injury to radiation workers during their lifetime.

The second method serves as the basis for establishing maximum permissible levels for the population at large, i. e., levels of radioactivity which, in the opinion of the Commission, will cause only a negligible incidence of leukaemia and genetic mutations. In this context, "negligible incidence" must be taken to mean an incidence of the same order as that of naturally occurring leukaemias and mutations.

In fixing maximum permissible levels of radiation, three types of data can serve as a basis: the levels of natural background radiation; the results of animal experiments; and observations in man.

(1) *Natural background radiation.* Man has always been exposed to background radiation from cosmic rays and from radionuclides present in soil (uranium, thorium), water (radium) and the atmosphere (radon), as well as in the interior of the human body (^{40}K and ^{14}C). It may be estimated that the world population is permanently exposed to a background radiation of between 100 and 150 mrem per year, with local or seasonal fluctuations. This dose may therefore serve as a basis for fixing maximum permissible levels for the general public.

(2) *Animal experiments.* Innumerable data on the somatic effects of ionizing radiations have been derived from animal experiments, particularly on mammals. Data have also been obtained on the induction of cancer and genetic mutations, but these results are more difficult to interpret, although they at least make it possible to define more and more accurately the dose capable of doubling the frequency of such events. Great caution must be exercised, however, in extrapolating these findings to man.

(3) *Observations in man.* Data derived from observations in man are of great importance. Such observations may be made on: patients treated by radiotherapy or radioisotopes; workers exposed to ionizing radiations or poisoned by radioactive substances, especially radium; and victims of the atomic bomb explosions at Hiroshima and Nagasaki. In many cases, the findings confirm the results of animal experiments.

Absolute values for maximum permissible doses

The International Commission on Radiological Protection has established maximum permissible doses of radiation which are valid for any type of exposure to external radiation or to radioactive contamination. These values are expressed in rem. They have been established initially for occupational exposure, taking account mainly of somatic effects. The maximum permissible weekly dose for a radiation worker has been fixed at 0.3 rem.

To allow for certain somatic risks (shortening of the life-span, induction of leukaemia), the maximum permissible total dose for a whole life-time has been fixed at 200 rem, corresponding to 5 rem per year or 0.1 rem per week for continuous exposure. Furthermore, to minimize the hazard of genetic effects, it is recommended that no worker should be exposed to more than 50 rem before the age of 30.

These values apply to continuous exposure. In the case of occasional exposure, a tolerance factor of 10 has been adopted, raising the maximum permissible dose to 3 rem, on condition that this dose is not exceeded for any 13 consecutive weeks.

These values apply to whole-body exposure. For partial exposure, a tolerance factor of 5 has been allowed, except in the case of certain particularly sensitive organs.

Finally, on the basis of observations made in cases of radium poisoning in man, it has been possible to establish directly that the maximum permissible body burden in the case of radium is 0.1 μc.

All these maximum permissible doses apply to occupational exposure. Where the population at large is concerned, greater stringency is recommended. If only a small fraction of the population is at risk, the values given above may be divided by 10, but if the whole population of a country, or a large part of it, is liable to exposure, it is recommended that the maximum level of artificial radioactivity should be of the same order as the natural background.

Practical values for maximum permissible doses

It is difficult in practice to measure radiation exposure directly in rem. The limits usually prescribed are therefore based on a different type of measurement, although they must, of course, respect absolutely the maximum permissible doses given above. These limits are derived by employing a factor known as the relative biological effectiveness, the basis of reference being an X-radiation of precisely defined characteristics. In the case of radioactive contamination, the determination of maximum permissible values is based on the following principles:

(1) The values are calculated with reference to a "standard man", i. e., an average man weighing 70 kg, having a body of precisely defined chemical composition, breathing 20 000 litres of air each day, ingesting 2½ litres of water each day, eliminating the same amount, and so on. Furthermore, for each radionuclide the critical organ or organs are determined, i. e., the tissues or organs irradiation of which carries the greatest risk to the health of the individual.

(2) In the case of internal contamination, the radiation to the critical organ from the absorbed radionuclide should not exceed the maximum permissible dose determined as described above and expressed in rem per unit of time. This applies to the vast majority of radionuclides, with the exception of bone-seeking alpha-emitters, for which the maximum

permissible body burden is determined from a direct comparison with radium. The maximum permissible body burdens of the principal radionuclides, established in this way, are expressed in μc.

(3) In the majority of cases, however, direct measurement of the concentration of radionuclides in the body is not possible. It is therefore necessary to establish maximum permissible concentrations for the environment, where the level of radioactive contamination can be measured directly. For this purpose, the level of internal contamination is considered as constant, a state of equilibrium being reached between absorption and elimination. It is possible to calculate the daily intake of radionuclides through the digestive or respiratory tract necessary to maintain this equilibrium. From this value, the maximum permissible concentrations of the radionuclide in drinking water or inhaled air can be deduced. This has been done for the principal radionuclides, the concentrations being expressed in μc/ml.

Acceptable limits for radioactive pollution of the atmosphere

The establishment of maximum permissible limits for radioactive pollution of the atmosphere is thus seen to be a special case of the general problem just discussed. For the principal radionuclides, maximum permissible concentrations in air, expressed in μc/ml, have been established by the International Commission on Radiological Protection. The table reproduced in the annex is based on recommendations adopted at a meeting held in Geneva in 1956. The following comments may be made regarding the establishment of these limits and the utilization of the table.

(1) In fixing these limits, certain assumptions were made, depending on the type of radioactive pollution.

In the case of pollution by rare gases (^{41}A) it is assumed that only external exposure to the surrounding cloud is involved, the whole body constituting the critical organ.

In the case of pollution by particulate matter, it is assumed that 25% of the particles are exhaled again, 50% deposited in the upper respiratory tract and subsequently swallowed, and 25% deposited in the lungs, from where, if the substances are soluble, they are absorbed into the body.

In the event of soluble radioactive substances being inhaled, it is assumed that the critical organ may be either the lungs, or an internal organ in which the radionuclide, after having been distributed throughout the body, accumulates selectively. If the inhaled radioactive substances are insoluble or only slightly soluble, the critical organ is assumed to be either the lungs or—after the particles have been swallowed—part of the gastro-intestinal tract.

(2) The maximum permissible concentration in air for each radionuclide has been established on the assumption that the atmospheric pollution is due solely to this radionuclide and is constant. The values will consequently have to be modified if several pollutants are present or if pollution is intermittent.

In the case of atmospheric pollution by several radionuclides, the total dose of radiation received by the body should not exceed the absolute value for the maximum permissible dose. To obtain the permissible concentration of each radionuclide, the figure given in the table must therefore be corrected by a factor corresponding to the relative contribution of that radionuclide to the total atmospheric pollution.

In many cases, the problem is not only that pollution is due to several radionuclides but also that the precise nature of the radionuclides and their relative proportions are unknown. In such circumstances, the following maximum permissible concentrations have been recommended:

for any mixture of beta-gamma-emitters with the exception of ^{90}Sr: 10^{-9} μc/ml;

for any mixture of alpha-emitters with the exception of ^{239}Pu and ^{227}Ac: 5.10^{-12} μc/ml.

If exposure is only occasional, it may be assumed that concentrations ten times the above values will not be harmful, provided that the average concentration during any period of 12 months does not exceed the recommended values.

(3) The maximum permissible concentrations given in the table are intended to apply to occupational exposure. In the case of the population at large, additional safety factors must be adopted. If only a small part of the population is exposed to the polluted atmosphere, the figures in the table should be divided by 10. If there is a possibility that the whole population, or a large part of it will be affected, the maximum permissible concentrations must be reduced to such levels that the resulting exposure is of the same order as that due to the natural background.

Monitoring and Prevention of Radioactive Pollution of the Atmosphere

Measures for preventing possible harmful effects of radioactive pollution of the atmosphere must be based on the fundamental principle that the prescribed maximum permissible doses should on no account be exceeded. The first requirement is therefore a rigorous monitoring system to determine the nature and the level of radioactive contamination; effective preventive methods can then be applied.

Monitoring of pollution

The aim of monitoring is to determine the type of pollutant present and the level of radioactivity. This enables an assessment to be made of the hazard run by radiation workers and by the general population and, at the same time, provides a check on the effectiveness of the measures employed to control the pollution.

General principles

Before discussing the techniques of monitoring, it is necessary to examine the principles upon which the organization and execution of such a programme of control of atmospheric pollution must be based.

(1) To increase the accuracy and reliability of the results, monitoring must be systematic and simultaneous cross-checks should be made at as many points as possible.

These precautions are necessary because the ordinary human senses give no warning of the presence of ionizing radiations, even in amounts that are distinctly toxic; at best they can detect the material vehicles of radioactive pollutants. Moreover, the pathological effects of exposure to radiations do not have any specific etiological characteristics and might be due to other causes; and, in addition, there may often be a considerable latent period before the harmful effects of exposure become manifest. It is therefore evident that the expert required to establish a cause-and-effect relationship in cases of suspected radiation injury is confronted with very intricate problems. Finally, the almost total lack of any means of preventing or curing the harmful effects of the inhalation of radioactive substances makes it imperative that there should be a rigorous system of monitoring to ensure the immediate detection of dangerous levels of radioactive pollution.

(2) Monitoring must provide information not only on the amount of radioactivity present but also on the nature of the pollutants, since, as has already been shown, wide variations in toxicity may exist between the various radionuclides. It should be made clear from the outset that both qualitative and quantitative analyses are extremely difficult.

From the quantitative aspect, the maximum permissible concentrations in air for many of the radionuclides are so small that the analyses have to be made at the limit of sensitivity of the most up-to-date equipment. If a large number of observations are needed and these highly refined techniques have to be employed as routine methods, it can be readily imagined that the technical problems are extremely difficult to solve.

On the qualitative side, the isolation and identification of the various radionuclides is very difficult when only traces are present. Even in the case of a single radionuclide, the task is by no means an easy one, and the difficulties increase with the number of radioactive substances present. In the case of very complex mixtures, separation may be impossible with the techniques at present available. As already indicated earlier, the solution to this problem is to assume that the mixture consists entirely of the most toxic radionuclide present. It should be noted that a knowledge of the working conditions often provides a useful pointer to the nature of the pollutants and enables the investigations to be considerably simplified.

(3) The monitoring system should be such that it enables all the sources of radioactive pollution of the atmosphere to be identified. Moreover,

since the aim is to ensure healthy working and living conditions, it must cover both occupational exposure and exposure of the population at large.

With regard to occupational exposure, several types of atmosphere have to be investigated: the air in the premises occupied by the workers; the air in confined spaces to which access is restricted because of permanent pollution; the air in confined spaces to which access is prohibited; gases or vapours which are discharged into the outside air, before and after filtration; and air in the neighbourhood of atomic installations. Tests should also be made to detect secondary surface contamination occurring as the result of atmospheric pollution, as well as skin or internal contamination of workers.

For the protection of the population at large, a monitoring system must be set up which will ensure the detection of radioactive pollution in the neighbourhood of any installations where the continuous or accidental release of radioactive substances into the atmosphere is liable to occur. In addition, there must be some national body responsible for measuring the general atmospheric pollution resulting from nuclear tests. These methods should be supplemented by suitable tests to determine the level of secondary contamination at various points in the food chain. Secondary contamination of the general population is only likely to arise in exceptional circumstances as the result of an accident.

Techniques of monitoring

The techniques involved in monitoring radioactive pollution of the atmosphere may be grouped under three headings: sampling, quantitative analysis, and qualitative analysis.

Sampling. It is rarely possible to measure directly the level of radioactive pollution of the atmosphere. This can be done, however, in the case of gaseous gamma-emitters, such as ^{41}A, simply by measuring the dose of gamma radiation.

In the vast majority of cases, analyses are performed on samples of air, which may be classified in various ways.

First of all, a distinction may be made between gross sampling and selective sampling. Gross samples are representative of the atmosphere as a whole and are obtained by admitting a quantity of air into an ionization chamber or scintillation counter. Selective samples contain only certain of the components of the atmosphere. The selection of these components may be made either by physical or by chemical methods. Physical methods are employed mainly for the detection of radioactive particulates. The method most frequently used consists in passing the air through a filter which retains only particles of a certain size. In another method used in general monitoring, the particles are collected by deposition on adhesive paper. Physicochemical methods can be applied to certain gases, such as radon, which is retained in good yield if the sample is passed over activated charcoal. Chemical selection depends on reactions capable of retaining the radioactive contaminants and is used especially for gases. For example, radioactive carbon dioxide can be fixed by bubbling the air through lime water.

Sampling may be done either on a continuous or on a periodic basis. The advantage of continuous sampling is that it provides more complete information on changes in atmospheric pollution at a given point over a period of time. Continuous sampling can be effected, for example, by passing the air through an ionization chamber or by continuous aspiration through a filter. In the majority of cases, however, periodic samples are taken. These are easier to obtain and the use of continuous recording equipment is unnecessary.

Quantitative analysis. After the samples have been collected, they must be analysed to determine the amounts of radionuclides present and hence the level of radioactive contamination of the atmosphere. Fundamentally, there are three types of method at present available for measuring radiation.

(1) *Electrometric methods.* These are based on the ionization produced when the radiation passes through a certain volume of gas subjected to a potential difference. If the potential difference is small, the result is simply the transfer of ions to one electrode with the production of a current which can be recorded; this is what happens in an ionization chamber. If the potential difference is sufficiently high, the acceleration of the ions formed leads to "avalanche" phenomena, culminating in an electric pulse which can be recorded; this is the principle of the discharge counter tube, which may be either of the proportional or of the Geiger-Müller type.

(2) *Scintillometric methods.* These methods depend on the fact that if a crystal of suitable material (sodium iodide for gamma rays, anthracene for beta rays, zinc sulfide for alpha rays) is exposed to ionizing radiation a bright flash of light is produced in the crystal. By a photoelectric effect this gives rise to an electron flux which is detected by a photomultiplier tube, amplified, and converted into an electric pulse which is recorded. The whole apparatus is known as a scintillation counter.

(3) *Photometric methods.* These are based on chemical changes appearing in an emulsion sensitive to ionizing radiations. The result may be either a uniform darkening of the film or the appearance of tracks following the path of strongly ionizing particles, such as alpha rays. For the former purpose, sensitive films are used, and for the latter, nuclear track-plates.

The final results obtained by any of these methods fall into two groups. Those in the first group (ionization chamber, sensitive film) are a reflection of statistical phenomena and measure the exposure dose in roentgens; those in the second group (Geiger-Müller counters, scintillation counters, nuclear track-plates) depend on the counting of single events and measure the particle flux or the activity in curies.

The results of these analytical determinations are not suitable for direct evaluation. The radiation doses or amounts of radionuclides must first be converted into radioactive concentrations per unit volume of air. In certain cases, e. g., with data obtained by ionization measurements, such a conversion is relatively easy, but it is much more difficult when the data are of the type furnished by counting methods, owing to the intervention of other factors (geometrical considerations, chance effects, etc.). Moreover, in general it is necessary to know the nature of the radioactive pollutant before the quantitative data can be interpreted satisfactorily.

Qualitative analysis. A knowledge of the working conditions and the hazards involved will usually provide an indication of the nature of the radionuclides likely to be present and thus facilitate qualitative analysis. The methods used are of two types, depending upon whether they are based on the physicochemical or radioactive properties of the pollutants.

Separation of the radionuclides and possibly their identification may be effected on the basis of their physical and chemical properties. The distinction between gaseous and particulate contaminants is easily made. The latter can very often be separated by granulometric analysis, a method that has the advantage of providing valuable information on the true toxicity of radioactive dusts.

Chemical methods of separation, such as co-precipitation and ion exchange, are particularly suitable for the analysis of samples of radioactive dusts. They permit not only the isolation of the radionuclides but also their identification. Considerable difficulties are experienced, however, as soon as the degree of pollution is insufficient to give weighable quantities, especially in the case of radionuclides that have no stable isotopes that can act as carriers.

Analytical methods based on the radioactive properties of the pollutants make use of their decay and emission characteristics. The half-life of a radionuclide is a specific constant by which it can be identified. The decay method consists in determining the amount of radioactive material present in the sample at a series of time intervals and hence calculating the half-life. This very simple method is effective if the half-life is short. By the application of mathematical analysis to the decay curve, it can also be used for mixtures provided that the number of components is small. It is useless, however, in the case of complex mixtures or of radionuclides with a long half-life. Methods must then be used which depend on differences in the nature and energy of the radiations emitted by the various radionuclides. The use of detectors having a specific sensitivity to alpha, beta or gamma rays—in particular, scintillation counters—enables a rough preliminary qualitative analysis to be made. With certain types of detector (e. g., proportional counters or nuclear track-plates) the radiations can be differentiated according to their energy. By means of "electronic discrimination", it is possible to single out those radiations having energies between certain limits and to perform a spectrographic analysis. In particular, the gamma spectrometer operating by amplitude selection is coming more and more into use for routine analysis. Finally, in the absorption method, screens of suitable material and thickness are employed which are capable of selective absorption of certain radiations, thus enabling the radiation or radiations to be identified.

Instrumentation

The performance of these qualitative and quantitative analyses calls for suitable instruments constructed in a wide range of materials. Generally speaking, the apparatus used for radiation detection must satisfy rigorous requirements, particularly with regard to sensitivity, accuracy, and robustness. No attempt will be made here to give even an incomplete list of the materials used, but, by way of illustration, a few detectors and their accessories will be briefly described.

For atmospheric sampling, simple bottles which have been previously evacuated may be used to collect gross samples, whereas selective samples are generally obtained by means of suction pumps of medium capacity, fitted with suitable filters and connected either to a power supply or to a battery. The detectors themselves are of many types. Depending upon the measurements to be made, ionization chambers and discharge tube counters vary in shape and size, in the nature of the gas they contain and the method by which the gas is generated, and in the thickness of the walls and the material from which they are constructed. Similarly, the crystals used in scintillation counters differ in composition, shape and size according to whether they are to be used for the detection of alpha, beta or gamma radiation. Finally, sensitive film and nuclear track-plates are manufactured in shapes which facilitate their exposure to the samples. The apparatus used for screen analysis and for chemical methods does not exhibit any special features. On the other hand, for spectrographic analysis of ionizing radiations, particularly gamma radiation, very complicated instruments are needed, capable of selecting particular energy bands. Such instruments may be either of the single-channel type with variable band or of the multichannel type with set bands enabling a rapid and complete analysis to be carried out.

To obtain certain results, a number of accessory instruments are necessary. Radiation detectors may be classed either as dosimeters or as rate-meters, depending upon whether they measure the quantity or the intensity of the radiation. The rate-meters are very interesting since they enable instantaneous measurements of radioactivity to be made, but they must be used in conjunction with recording devices if it is desired to follow the changes in radioactive pollution over a period of time. They may also be coupled with auditory or visual alarm signals which are actuated whenever the maximum permissible concentration in air is exceeded.

The equipment necessary for detecting radioactive pollution of the air thus comprises a whole range of detectors. The simplest are mobile and provide information on pollution at a given point and a given time, while the more complicated ones are fixed and effect instantaneous measurements, continuous recording or, if necessary, qualitative analysis of air pollution.

Applications

The choice of techniques and instruments will depend on the type of work being performed and on the nature of the resulting air pollution. Only a few examples can be given here to illustrate the applications of the above techniques and the usefulness of the methods described.

One of the best examples of local atmospheric pollution is found in the uranium mines, where the pollutants are natural radioactive materials in gaseous or particulate form. The monitoring technique at present in use consists in systematic sampling of the air at representative points in the galleries in accordance with the type of work in progress (drilling, blasting, removal of the ore, etc.). The bottles containing the crude samples are then connected with an ionization chamber, scintillation counter, or counter containing activated charcoal to measure the concentrations of radon. Selective sampling with the aid of suitable filters is employed to study the particle size and chemical composition of dusts, as well as to measure their radioactivity, for which purpose nuclear track-plates are particularly useful. For special studies, continuous sampling apparatus may be used, enabling a continuous record to be made of atmospheric radioactivity.

An example of regional pollution is to be found in the neighbourhood of atomic installations. The chimneys of these installations continuously pollute the atmosphere with radioactive gases emanating from leaks in the coolant system of the reactor; occasionally, they may also emit radioactive gases or dusts resulting from fires or other accidents in the reactor. Monitoring is effected by a chain of stations suitably arranged around the site, their position depending upon meteorological factors and demographic considerations. These stations should make possible the continuous monitoring of radioactive contamination of the atmosphere. Radiation from ^{41}A is measured by means of coupled (open and closed) ionization chambers. By using selective sampling equipment in which the air is passed through a band of filter paper, which is continuously unrolled and then travels in front of a discharge or scintillation counter, it is possible to measure pollution with radioactive dusts. Ionization chambers and filter detectors give instantaneous information on pollution with radioactive gases and dusts; when used in conjunction with recording equipment they enable the average pollution at the point considered to be determined; when fitted with alarm devices, they can give a warning if the maximum permissible concentrations are exceeded. Finally, in the event of an accident, subsequent determination of the particle size of the material retained on the filter, or its analysis by chemical or radiochemical methods, will indicate the nature of the contaminants.

The apparatus just described would also be employed for monitoring general atmospheric pollution arising from nuclear tests. However, for measurements on a very large scale, the most usual procedure consists in allowing the radioactive particles to settle on adhesive paper exposed in the open air for a given time. The paper is ashed and from the activity of the residue measured with a counter, the radioactive fall-out can be determined in μc per unit of surface area. Qualitative analysis is also possible in certain circumstances, particularly for the identification of ^{90}Sr.

Supplementary surveys

For a really complete assessment of all the consequences to man and his environment of radioactive pollution of the atmosphere, the monitoring programmes described above should be supplemented by surveys to determine the secondary contamination of the environment and of man himself. Here it is only possible to outline the general methods used in these two types of survey, since a comprehensive treatment would take us too far from the subject of atmospheric pollution.

Environmental surveys. It has already been shown that atmospheric pollution leads, by natural deposition of the pollutants, to contamination of the soil, water, flora and fauna and thus to contamination of the food chain. Monitoring of such contamination cannot be done on a systematic basis but only in the form of spot checks, i. e., tests made on samples taken at suitably chosen times and places. In the case of soil, flora and fauna, for example, samples are collected in the immediate vicinity of atomic installations and to the lee of the prevailing wind; and in the case of food, tests are made on those foods likely to introduce into the body the largest quantities of contaminants (e. g., in European countries, milk is the principal vector of ^{90}Sr). The very small quantities of radionuclides present in these samples make quantitative measurements very difficult, especially as they are dispersed in a very large mass of inert material. Self-absorption therefore becomes a highly important factor, a difficulty not experienced in the case of atmospheric pollution. Consequently, the samples must be given a preliminary treatment, the amount of inert matter being reduced to a minimum by desiccation and conversion to inorganic salts. The techniques for the measurement of gamma radiation present fewer difficulties. In the case of contamination by beta emitters, it is necessary to work with very thin samples and to use highly sensitive counters. For alpha emitters, it is preferable to use counters into which the sample can be introduced in gaseous form. Special, highly sensitive equipment has been devised for the direct measurement of gamma radiation on intact biological samples. The difficulty that remains is that of the background counting rate of these instruments, since the activity to be measured is often lower than the natural background.

Qualitative analyses are carried out using the physicochemical and radiochemical methods already discussed.

Surveys of human contamination. To evaluate more precisely the direct or indirect sequelae of radioactive pollution of the atmosphere, surveys of human contamination are necessary. Systematic surveys should be organized among workers continually exposed to the risk of contamination via the respiratory tract, i. e., uranium miners, metallurgical workers handling uranium and plutonium, etc. In the case of other radiation workers, such surveys need only be made occasionally, and surveys of the general public will rarely be necessary except in the case of accidents resulting in a high level of pollution. The present practice is to make a certain number of post-mortem examinations to obtain an estimate of the level of contamination in the population at large.

In the case of occupational exposure, skin contamination resulting from atmospheric pollution is easily measured using scintillation counters sensitive to alpha, beta and gamma radiation. On the other hand, it is very difficult to make direct determinations of internal contamination. At a few installations in various parts of the world, extremely sensitive, heavily shielded scintillation counters are available which are capable of making qualitative and quantitative analyses of internal contamination by gamma-emitters. Such methods are not at present susceptible of routine use, however.

It is necessary, therefore, to rely on indirect methods. Nasal samples taken at the end of the working day are useful for the detection, but not for the measurement, of radioactive contamination of the nasal fossae; their main value lies in the fact that they can provide a timely warning of the need for more thorough investigation. Methods similar to those used for the determination of atmospheric radon can be employed to measure the radon in exhaled air, provided that, before the examination, the subject has been made to breathe radon-free air. The methods most used, however, for the evaluation of human contamination depend on the measurement of radioactive levels in excreta. In particular, radiotoxicological analyses of urine can be used for the routine estimation of the most usual radioactive contaminants (uranium, radium, plutonium, strontium, caesium, etc.) and are sufficiently sensitive to detect amounts corresponding to the maximum permissible levels. The first stage in these analytical procedures consists in the isolation by chemical methods of the radionuclide or radionuclides present in the sample. It is therefore useful to have information on the nature of the radioactive pollution of the atmosphere in which the subject has been working. The activity of the sample obtained after chemical separation is estimated by the counting methods already described. There are two points to be noted; (*a*) this technique has so far been perfected only for the evaluation of contamination by a small number of radionuclides; and (*b*) the results obtained, expressed in μc per litre of urine, do not allow a direct assessment to be made of the level of internal contamination, since the rate of excretion of radionuclides is subject to considerable fluctuations, depending on a variety of factors. It is only by repeating such analyses at frequent intervals that the gravity of the situation can be properly assessed in cases of heavy contamination.

With regard to the general public, radiotoxicological analyses of this type will only be contemplated in the case of serious accidents involving whole populations—for example, those living in the neighbourhood of a nuclear reactor. As already mentioned, estimates of human contamination resulting from general pollution of the atmosphere are at present based on post-mortem examination, particular attention being paid to the presence of ^{90}Sr and ^{137}Cs.

Prevention of atmospheric pollution

It has already been pointed out that radioactive contamination cannot be detected by the human senses; that a considerable period may elapse between the time of exposure and the manifestation of the pathological effects; and that no really effective therapy is known for preventing or curing the harmful effects of internal contamination. It follows that for health protection reliance must be placed mainly on measures for ensuring that the radioactive pollution of the atmosphere does not reach dangerous levels.

General principles

Methods for the prevention of atmospheric pollution by radioactive substances are based on a number of general principles of which the most important are the following:

(1) For the reasons indicated above, it is imperative that the level of radioactive pollution of the atmosphere should not be allowed to exceed the maximum permissible limits. The measures adopted must therefore be particularly rigorous; they must not only be systematic, they must also be organized in such a way as to provide multiple and successive safeguards. Moreover, the organization must take account not only of the risks of continuous pollution arising out of normal working conditions, but also of the risks of accidental pollution due to defective installations.

(2) The radioactivity of a substance is a phenomenon that man is at present incapable of modifying in any way whatsoever. The rate of decay of radioactive substances and the nature and energy of the radiation emitted during the process remain completely unaffected by any form of treatment. Unlike most chemical pollutions, therefore, the pollutants cannot be converted into non-toxic substances; their rate of disappearance will depend

solely on the decay constant. In the case of radionuclides with a short half-life (^{24}Na, ^{32}P), the rate of spontaneous disappearance will be relatively rapid, but it will be almost negligible for elements with a long half-life (^{235}U, ^{239}Pu, ^{90}Sr, etc.). Attention must therefore be concentrated on the material vehicle of the radioactivity, except in the case of radionuclides with a very short half-life. Consequently, the preventive measures discussed below follow the same lines as those employed against atmospheric pollution in general, whether the contaminants are in gaseous or in particulate form.

(3) The preventive measures must be directed against both occupational exposure and exposure of the population at large. To be most effective, they should be organized primarily on a collective basis, i. e., the limitation of atmospheric pollution should be achieved by measures applied to the installations themselves. In certain cases, collective measures may prove inadequate, either because they are not technically feasible or because of accidents. They must therefore be supplemented by protective measures which can be applied on an individual basis. It is unnecessary to stress that such measures are highly inconvenient and that the security they afford is frequently insufficient. All the greater, therefore, is the obligation to develop measures of collective protection that are really efficacious.

General methods of prevention

To be effective, the control of radioactive pollution must follow a logical plan. It must comprise the following measures: limitation of the emission of radioactive pollutants; containment, to prevent, as far as possible, the spread of the pollution; and dispersal, to reduce the pollution below the maximum permissible level.

Limitation of the emission of radioactive pollutants. This is not only the most certain method of controlling radioactive pollution of the atmosphere, it is often the most economical as well. The measures to be adopted must be decided upon at a very early stage in the planning of installations, before construction commences. There is often a choice of several techniques for carrying out a particular operation, some of which offer particular advantages from the point of view of limiting air pollution. In uranium mines, for example, pollution can be kept to a minimum by the use of wet drilling, by underground drainage, and by clearing away the ore as rapidly as possible to prevent the release of radon. In the case of nuclear reactors, the risk of pollution can be reduced by the use of closed-cycle coolant systems containing gaseous coolants of the highest possible purity in order to avoid extraneous activation products. In the chemical and metallurgical industries, handling of the radioactive substances in the gaseous and powder forms should be kept to a minimum and metal working should be carried out under a jet of oil or water. Finally, in the case of nuclear tests, meteorological conditions must be such as to ensure minimum dispersal.

Containment. Measures aimed at preventing the spread of the material vector of radioactivity are of two types: containment of the polluted atmosphere and containment of the radioactive pollutant alone. In the first case, the aim is to separate the polluted atmosphere from the atmosphere of the area where people are working, or from the outside atmosphere. The separation may be imperfect, the exchange between polluted and unpolluted air being merely reduced to a minimum, as in the case of fume hoods. It is often an advantage, however, to ensure complete containment of the polluted air, as can be done with hermetically sealed boxes and closed-cycle systems. Selective methods for isolating the radioactive contaminants only usually depend on the use of filters which hold back radioactive particles of a given size. These filters must be able to resist corrosion or chemical attack by the radioactive substances. Provision must also be made for their replacement and decontamination without risk from the high concentration of radioactive material which they retain. For these reasons the filters are often arranged in series or in parallel.

Dispersal. It is not always possible by means of the two preceding methods to reduce radioactive pollution below the maximum permissible levels. If the atmospheric pollution is exceptionally high, methods of radioactive decontamination must be adopted. Only in the case of the short-lived pollutants is it possible to obtain satisfactory results by relying on spontaneous radioactive decay. In all other cases, measures must be adopted for dispersing the material vehicle of the radioactivity until concentrations are obtained which are lower than the prescribed maximum permissible limits. The dispersal method thus consists merely in spreading the pollution over a sufficiently large volume of air.

In the case of buildings where radiation work is performed, this is achieved mainly by adequate ventilation to ensure that the air inside the buildings is renewed and the radioactive contaminants are carried outside and dispersed; this is tantamount to a forced exchange between the polluted and the unpolluted atmosphere. It has not so far been found possible to make any appreciable use of physical, chemical, or electrostatic methods for radioactive decontamination of the atmosphere.

With regard to the population at large, the dispersal of radioactive pollutants is effected by the traditional use of high chimneys. Satisfactory dispersal to give concentrations below the maximum permissible limits depends on two sets of factors—on the position, height, and power of discharge of the chimney, and on local meteorological factors and the distribution of the population.

Additional protective measures

The measures described above are designed to reduce the level of radioactive pollution of the atmosphere below the maximum permissible limits.

In the vast majority of cases, this result is attainable provided that the installations function normally. However, in certain types of work involving very heavy pollution (uranium mining, metallurgical processing) and in the case of serious accidents, it may be necessary to provide additional protective measures, from the point of view both of environmental and of human contamination.

Environmental contamination. This is essentially a question of siting atomic installations and centres handling radioactive substances in such a way that the consequences of heavy atmospheric pollution will not be too serious. In choosing a suitable site, the most important considerations are the following:

(1) Type of installation, e. g., nuclear reactor, chemical treatment plant, plutonium extraction centre, etc. This will influence the type of accident most likely to occur. Moreover, the concentration of the buildings and the safety devices with which they are provided will profoundly affect the decisions taken regarding the choice of site. Thus, certain reactors used for research purposes present practically no hazard, whereas the hazards associated with some types of production reactor may be considerable.

(2) Propagation of radioactive contamination. Here the principal factors involved are meteorological, especially the local weather conditions: prevailing winds, rainfall pattern, temperature changes, humidity, etc.

(3) Nature of the environment liable to be contaminated, e. g., distribution of the population, position of industrial and residential areas and of agricultural zones, etc.

The choice of a suitable site will depend on a careful weighing up of all these considerations. The same arguments may also apply in part to the selection of sites for nuclear tests.

Human contamination. In spite of all the protective measures already discussed, atmospheric pollution may be of such a nature that steps have to be taken to provide individual protection for all exposed persons. The measures adopted may be of two very different types, depending upon whether they are aimed at the prevention or the treatment of contamination.

With rare exceptions, prevention of individual contamination is attempted only in cases of occupational exposure. Measures should be aimed at limiting both skin contamination and internal contamination via the respiratory tract. The first requirement is to encourage proper working habits, e. g., tidiness, no smoking, eating or drinking in contaminated rooms, and personal hygiene (washing the hands, taking showers). The risk of skin contamination can be reduced by wearing suitable working clothes (overalls, boots, caps, gloves, etc.). Contamination via the respiratory tract may be prevented by providing each worker with a suitable mask, hood, or protective suit giving selective or complete isolation from the radioactive pollutants. The inconvenience of wearing such protective clothing while working is a good argument for not using it except in conditions where it is absolutely impossible to achieve satisfactory results by collective measures. As already stated, such conditions may be found in highly contaminated mine workings or in atomic installations contaminated as the result of serious accidents.

The treatment of radioactive contamination of the skin is relatively easy. On the other hand, internal contamination resulting from the inhalation of radioactive substances is extremely difficult to treat owing to the fact that, in most cases, it is impossible to modify the metabolism of the contaminants to any appreciable extent. If the radionuclides are short-lived, the hazard is not great, except in the case of very heavy contamination. On the other hand, the fixation in the tissues of radionuclides having long radioactive and biological half-lives may have extremely serious consequences. The lack of any satisfactory treatment for poisoning by radioactive substances is one more argument for building up a really effective organization for the prevention of radioactive pollution of the atmosphere.

Conclusion

The rapidly expanding applications of radioactive materials and of nuclear energy must inevitably lead to a vast increase in radioactive pollution of the atmosphere unless effective preventive measures are taken without delay. Such measures, which are essential for safeguarding the health of both radiation workers and the population at large, must be conceived as part of a programme for the control of air pollution in general.

The first essential is an effective system of monitoring, the principles of which have been outlined above. The International Commission on Radiological Protection issues periodically recommendations regarding maximum permissible doses of radiation and maximum permissible concentrations of radionuclides in air, drinking water, etc. Similar recommendations are drawn up by national bodies responsible for advising governments on radiological protection. The monitoring system must be adequate to ensure that prompt action is taken whenever there is a danger of these maximum permissible levels being exceeded. Ultimately, the recommendations may be embodied in appropriate legislature, but caution is necessary at present in view of the uncertainty still surrounding the precise health hazards of a given exposure to radiation.

As soon as possible, efforts should be made to secure international agreement regarding methods of monitoring and control—all the more so as a source of radioactive pollution in one country may affect neighbouring territories. The World Health Organization can help to encourage such co-operation and can assist governments by the dissemination of the most recent information on the true hazards of radioactive pollution of the atmosphere and on available methods of monitoring, control, and radiation protection.

There is an urgent need for basic research in radiotoxicology and radiopathology in order that radiation hazards can be defined more precisely and radiation injuries diagnosed at an early stage and effectively treated. More sensitive and reliable techniques of radiochemical analysis are also needed, especially for the detection and identification of small quantities of radioactive contaminants.

If these problems are not tackled energetically, the whole future of the exploitation of nuclear energy for peaceful purposes will be jeopardized. Public opinion has already been aroused to the dangers of radioactive pollution of the atmosphere and the need for rigorous control. Public health authorities should grasp this opportunity to ensure that the potential benefits of nuclear energy can be enjoyed to the full without endangering the health of workers in this new industry or of the general population.

Annex

MAXIMUM PERMISSIBLE CONCENTRATIONS OF RADIONUCLIDES IN AIR FOR AN OCCUPATIONAL EXPOSURE OF 168 HOURS PER WEEK *

Radionuclide and type of decay		Critical organ **	Maximum permissible concentration in air (μc/ml)	Radionuclide and type of decay		Critical organ **	Maximum permissible concentration in air (μc/ml)
$_{1}H^{3}$ (HTO or H_2^3O) β^-	(sol.)	Body tissue Total body	2×10^{-6} 3×10^{-6}	$_{11}Na^{24}$ β^-, γ	(sol.)	GI (SI)	4×10^{-7}
(H_2^3)	(submersion)	Skin	4×10^{-4}		(insol.)	GI (LLI)	5×10^{-8}
$_{4}Be^{7}$ ε, γ	(sol.)	GI (LLI) Total body	4×10^{-6} 2×10^{-6}	$_{14}Si^{31}$ β^-, γ	(sol.)	GI (S)	2×10^{-6}
	(insol.)	Lung GI (LLI)	4×10^{-7} 3×10^{-6}		(insol.)	GI (ULI)	3×10^{-7}
$_{6}C^{14}(CO_2)$ β^-	(sol.)	Fat	10^{-6}	$_{15}P^{32}$ β^-	(sol.)	Bone	2×10^{-8}
	(submersion)	Total body	10^{-5}		(insol.)	Lung GI (LLI)	3×10^{-8} 4×10^{-8}
$_{9}F^{18}$ β^+	(sol.)	GI (SI)	2×10^{-6}	$_{16}S^{35}$ β^-	(sol.)	Testis	9×10^{-8}
	(insol.)	GI (ULI)	9×10^{-7}		(insol.)	Lung GI (LLI)	9×10^{-8} 5×10^{-7}
$_{11}Na^{22}$ β^+, γ	(sol.)	Total body	6×10^{-8}	$_{17}Cl^{36}$ β^-	(sol.)	Total body	10^{-7}
	(insol.)	Lung GI (LLI)	3×10^{-9} 5×10^{-8}		(insol.)	Lung GI (LLI)	8×10^{-9} 10^{-7}

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* Condensed from: International Commission on Radiological Protection (1959) *Recommendations of the International Commission on Radiological Protection. Report of Committee II on permissible dose for internal radiation*, London, New York, Paris, Los Angeles, Pergamon Press, pp. 41-84.

** The abbreviations GI, S, SI, ULI and LLI refer to gastrointestinal tract, stomach, small intestine, upper large intestine, and lower large intestine, respectively.

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{17}Cl^{38}$ β^-, γ	(sol.)	GI (S)	9×10^{-7}
	(insol.)	GI (S)	7×10^{-7}
$_{18}A^{37}$ ε	(submersion)	Skin	10^{-3}
$_{18}A^{41}$ β^-, γ	(submersion)	Total body	4×10^{-7}
$_{19}K^{42}$ β^-, γ	(sol.)	GI (S)	7×10^{-7}
	(insol.)	GI (LLI)	4×10^{-8}
$_{20}Ca^{45}$ β^-	(sol.)	Bone	10^{-8}
	(insol.)	Lung GI (LLI)	4×10^{-8} 3×10^{-7}
$_{20}Ca^{47}$ β^-, γ	(sol.)	Bone	6×10^{-8}
	(insol.)	GI (LLI) Lung	6×10^{-8} 6×10^{-8}
$_{21}Sc^{46}$ β^-, γ	(sol.)	GI (LLI) Liver	8×10^{-8} 8×10^{-8}
	(insol.)	Lung GI (LLI)	8×10^{-9} 7×10^{-8}
$_{21}Sc^{47}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	GI (LLI)	2×10^{-7}
$_{21}Sc^{48}$ β^-, γ	(sol.)	GI (LLI)	6×10^{-8}
	(insol.)	GI (LLI)	5×10^{-8}
$_{23}V^{48}$ $\beta^+, \varepsilon, \gamma$	(sol.)	GI (LLI)	6×10^{-8}
	(insol.)	Lung GI (LLI)	2×10^{-8} 5×10^{-8}
$_{24}Cr^{51}$ ε, γ	(sol.)	GI (LLI) Total body	4×10^{-6} 4×10^{-6}
	(insol.)	Lung GI (LLI)	8×10^{-7} 3×10^{-6}
$_{25}Mn^{52}$ $\beta^+, \varepsilon, \gamma$	(sol.)	GI (LLI)	7×10^{-8}
	(insol.)	Lung GI (LLI)	5×10^{-8} 5×10^{-8}
$_{25}Mn^{54}$ ε, γ	(sol.)	GI (LLI) Liver	3×10^{-7} 10^{-7}
	(insol.)	Lung GI (LLI)	10^{-8} 2×10^{-7}
$_{25}Mn^{56}$ β^-, γ	(sol.)	GI (LLI)	3×10^{-7}
	(insol.)	GI (LLI)	2×10^{-7}
$_{26}Fe^{55}$ ε	(sol.)	Spleen	3×10^{-7}
	(insol.)	Lung GI (LLI)	3×10^{-7} 4×10^{-6}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{26}Fe^{59}$ β^-, γ	(sol.)	GI (LLI) Spleen	10^{-7} 5×10^{-8}
	(insol.)	Lung GI (LLI)	2×10^{-8} 9×10^{-8}
$_{27}Co^{57}$ ε, γ, e^-	(sol.)	GI (LLI)	10^{-6}
	(insol.)	Lung GI (LLI)	6×10^{-8} 7×10^{-7}
$_{27}Co^{58m}$ β^+, ε, γ	(sol.)	GI (LLI)	6×10^{-6}
	(insol.)	Lung GI (LLI)	3×10^{-6} 4×10^{-6}
$_{27}Co^{58}$ β^+, ε	(sol.)	GI (LLI) Total body	3×10^{-7} 3×10^{-7}
	(insol.)	Lung GI (LLI)	2×10^{-8} 2×10^{-7}
$_{27}Co^{60}$ β^-, γ	(sol.)	GI (LLI) Total body	10^{-7} 10^{-7}
	(insol.)	Lung GI (LLI)	3×10^{-9} 6×10^{-8}
$_{28}Ni^{59}$ ε	(sol.)	Bone	2×10^{-7}
	(insol.)	Lung GI (LLI)	3×10^{-7} 3×10^{-6}
$_{28}Ni^{63}$ β^-	(sol.)	Bone	2×10^{-8}
	(insol.)	Lung GI (LLI)	10^{-7} 10^{-6}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{28}Ni^{65}$ β^-, γ	(sol.)	GI (ULI)	3×10^{-7}
	(insol.)	GI (ULI)	2×10^{-7}
$_{29}Cu^{64}$ β^-, β^+, ε	(sol.)	GI (LLI)	7×10^{-7}
	(insol.)	GI (LLI)	4×10^{-7}
$_{30}Zn^{65}$ β^+, ε, γ	(sol.)	Total body Prostate Liver	4×10^{-8} 4×10^{-8} 5×10^{-8}
	(insol.)	Lung GI (LLI)	2×10^{-8} 3×10^{-7}
$_{30}Zn^{69m}$ γ, e^-, β^-	(sol.)	GI (LLI) Prostate	2×10^{-7} 10^{-7}
	(insol.)	GI (LLI)	10^{-7}
$_{30}Zn^{69}$ β^-	(sol.)	GI (S) Prostate	4×10^{-6} 2×10^{-6}
	(insol.)	GI (S)	3×10^{-6}
$_{31}Ga^{72}$ β^-, γ	(sol.)	GI (LLI)	8×10^{-8}
	(insol.)	GI (LLI)	6×10^{-8}
$_{32}Ge^{71}$ ε	(sol.)	GI (LLI)	4×10^{-6}
	(insol.)	Lung GI (LLI)	2×10^{-6} 3×10^{-6}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{33}\text{As}^{73}$ ε, γ	(sol.)	GI (LLI) Total body	10^{-6} 7×10^{-7}
	(insol.)	Lung GI (LLI)	10^{-7} 8×10^{-7}
$_{33}\text{As}^{74}$ β^-, β^+, ε, γ	(sol.)	GI (LLI)	10^{-7}
	(insol.)	Lung GI (LLI)	4×10^{-8} 9×10^{-8}
$_{33}\text{As}^{76}$ β^-, γ	(sol.)	GI (LLI)	4×10^{-8}
	(insol.)	GI (LLI)	3×10^{-8}
$_{33}\text{As}^{77}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	GI (LLI)	10^{-7}
$_{34}\text{Se}^{75}$ ε, γ	(sol.)	Kidney Total body	4×10^{-7} 5×10^{-7}
	(insol.)	Lung GI (LLI)	4×10^{-8} 5×10^{-7}
$_{35}\text{Br}^{82}$ β^-, γ	(sol.)	Total body GI (SI)	4×10^{-7} 6×10^{-7}
	(insol.)	GI (LLI)	6×10^{-8}
$_{36}\text{Kr}^{85m}$ β^-, γ	(submersion)	Total body	10^{-6}
$_{36}\text{Kr}^{85}$ β^-	(submersion)	Total body	3×10^{-6}
$_{36}\text{Kr}^{87}$ β^-, γ	(submersion)	Total body	2×10^{-7}
$_{37}\text{Rb}^{86}$ β^-, γ	(sol.)	Total body Pancreas Liver	10^{-7} 10^{-7} 10^{-7}
	(insol.)	Lung GI (LLI)	2×10^{-8} 4×10^{-8}
$_{37}\text{Rb}^{87}$ β	(sol.)	Pancreas Total body Liver	2×10^{-7} 2×10^{-7} 2×10^{-7}
	(insol.)	Lung GI (LLI)	2×10^{-8} 3×10^{-7}
$_{38}\text{Sr}^{85m}$ ε, γ	(sol.)	GI (SI)	10^{-5}
	(insol.)	GI (SI)	10^{-5}
$_{38}\text{Sr}^{85}$ ε, γ	(sol.)	Total body	8×10^{-8}
	(insol.)	Lung GI (LLI)	4×10^{-8} 3×10^{-7}
$_{38}\text{Sr}^{89}$ β^-	(sol.)	Bone	10^{-8}
	(insol.)	Lung GI (LLI)	10^{-8} 5×10^{-8}
$_{38}\text{Sr}^{90}$ β^-	(sol.)	Bone	10^{-10}
	(insol.)	Lung GI (LLI)	2×10^{-9} 6×10^{-8}

Radionuclide and type of decay	Critical organ	Maximum permissible concentration in air (μc/ml)
$_{38}\mathrm{Sr}^{91}$ (sol.) β^-, γ	GI (LLI)	2×10^{-7}
(insol.)	GI (LLI)	9×10^{-8}
$_{38}\mathrm{Sr}^{92}$ (sol.) β^-, γ	GI (ULI)	2×10^{-7}
(insol.)	GI (ULI)	10^{-7}
$_{39}\mathrm{Y}^{90}$ (sol.) β^-	GI (LLI)	4×10^{-8}
(insol.)	GI (LLI)	3×10^{-8}
$_{39}\mathrm{Y}^{91m}$ (sol.) β^-, γ	GI (SI)	8×10^{-6}
(insol.)	GI (SI)	6×10^{-6}
$_{39}\mathrm{Y}^{91}$ (sol.) β^-, γ	GI (LLI) Bone	6×10^{-8} 10^{-8}
(insol.)	Lung GI (LLI)	10^{-8} 5×10^{-8}
$_{39}\mathrm{Y}^{92}$ (sol.) β^-, γ	GI (ULI)	10^{-7}
(insol.)	GI (ULI)	10^{-7}
$_{39}\mathrm{Y}^{93}$ (sol.) β^-, γ, e^-	GI (LLI)	6×10^{-8}
(insol.)	GI (LLI)	5×10^{-8}
$_{40}\mathrm{Zr}^{93}$ (sol.) β^-, γ, e^-	GI (LLI) Bone	2×10^{-6} 4×10^{-8}
(insol.)	Lung GI (LLI)	10^{-7} 10^{-6}

Radionuclide and type of decay	Critical organ	Maximum permissible concentration in air (μc/ml)
$_{40}\mathrm{Zr}^{95}$ (sol.) β^-, γ, e^-	GI (LLI) Total body	10^{-7} 4×10^{-8}
(insol.)	Lung GI (LLI)	10^{-8} 10^{-7}
$_{40}\mathrm{Zr}^{97}$ (sol.) β^-, γ	GI (LLI)	4×10^{-8}
(insol.)	GI (LLI)	3×10^{-8}
$_{41}\mathrm{Nb}^{93m}$ (sol.) γ, e^-	GI (LLI) Bone	9×10^{-7} 4×10^{-8}
(insol.)	Lung GI (LLI)	5×10^{-8} 7×10^{-7}
$_{41}\mathrm{Nb}^{95}$ (sol.) β^-, γ	GI (LLI) Total body	2×10^{-7} 2×10^{-7}
(insol.)	Lung GI (LLI)	3×10^{-8} 2×10^{-7}
$_{41}\mathrm{Nb}^{97}$ (sol.) β^-, γ	GI (ULI)	2×10^{-6}
(insol.)	GI (ULI)	2×10^{-6}
$_{42}\mathrm{Mo}^{99}$ (sol.) β^-, γ	Kidney GI (LLI)	3×10^{-7} 5×10^{-7}
(insol.)	GI (LLI)	7×10^{-8}
$_{43}\mathrm{Tc}^{96m}$ (sol.) ε, γ, e^-	GI (LLI)	3×10^{-5}
(insol.)	Lung GI (LLI)	10^{-5} 2×10^{-5}

Radionuclide and type of decay	Critical organ	Maximum permissible concentration in air ($\mu c/ml$)	Radionuclide and type of decay	Critical organ	Maximum permissible concentration in air ($\mu c/ml$)
${}_{43}\mathrm{Tc}^{96}$ (sol.) ε, γ	GI (LLI)	2×10^{-7}	${}_{44}\mathrm{Ru}^{105}$ (sol.) β^-, γ, e^-	GI (ULI)	2×10^{-7}
(insol.)	GI (LLI)	8×10^{-8}	(insol.)	GI (ULI)	2×10^{-7}
${}_{43}\mathrm{Tc}^{97m}$ (sol.) ε, γ, e^-	GI (LLI)	8×10^{-7}	${}_{44}\mathrm{Ru}^{106}$ (sol.) β^-, γ	GI (LLI)	3×10^{-8}
(insol.)	Lung GI (LLI)	5×10^{-8} 3×10^{-7}	(insol.)	Lung GI (LLI)	2×10^{-9} 2×10^{-8}
${}_{43}\mathrm{Tc}^{97}$ (sol.) ε	GI (LLI) Kidney	4×10^{-6} 4×10^{-6}	${}_{45}\mathrm{Rh}^{103m}$ (sol.) γ, e^-	GI (S)	3×10^{-5}
(insol)	Lung GI (LLI)	10^{-7} 10^{-6}	(insol.)	GI (S)	2×10^{-5}
${}_{43}\mathrm{Tc}^{99m}$ (sol.) β^-, γ	GI (ULI)	10^{-5}	${}_{45}\mathrm{Rh}^{105}$ (sol.) β^-, γ	GI (LLI)	3×10^{-7}
(insol.)	GI (ULI)	5×10^{-6}	(insol.)	GI (LLI)	2×10^{-7}
${}_{43}\mathrm{Tc}^{99}$ (sol.) β^-	GI (LLI)	7×10^{-7}	${}_{46}\mathrm{Pd}^{103}$ (sol.) ε, γ, e^-	GI (LLI) Kidney	8×10^{-7} 5×10^{-7}
(insol.)	Lung GI (LLI)	2×10^{-8} 3×10^{-7}	(insol.)	Lung GI (LLI)	3×10^{-7} 5×10^{-7}
${}_{44}\mathrm{Ru}^{97}$ (sol.) ε, γ, e^-	GI (LLI)	8×10^{-7}	${}_{46}\mathrm{Pd}^{109}$ (sol.) β^-, γ, e^-	GI (LLI)	2×10^{-7}
(insol.)	GI (LLI) Lung	6×10^{-7} 7×10^{-7}	(insol.)	GI (LLI)	10^{-7}
${}_{44}\mathrm{Ru}^{103}$ (sol.) β^-, γ, e^-	GI (LLI)	2×10^{-7}	${}_{47}\mathrm{Ag}^{105}$ (sol.) ε, γ	GI (LLI)	2×10^{-7}
(insol.)	Lung GI (LLI)	3×10^{-8} 10^{-7}	(insol.)	Lung GI (LLI)	3×10^{-8} 2×10^{-7}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{47}\mathrm{Ag}^{110m}$ β^-, γ	(sol.)	GI (LLI)	7×10^{-8}
	(insol.)	Lung GI (LLI)	3×10^{-9} 5×10^{-8}
$_{47}\mathrm{Ag}^{111}$ β^-, γ	(sol.)	GI (LLI)	10^{-7}
	(insol.)	GI (LLI)	8×10^{-8}
$_{48}\mathrm{Cd}^{109}$ ε, γ, e^-	(sol.)	GI (LLI) Liver Kidney	4×10^{-7} 2×10^{-8} 2×10^{-8}
	(insol.)	Lung GI (LLI)	3×10^{-8} 3×10^{-7}
$_{48}\mathrm{Cd}^{115m}$ β, γ, e^-	(sol.)	GI (LLI) Liver Kidney	6×10^{-8} 10^{-8} 2×10^{-8}
	(insol.)	Lung GI (LLI)	10^{-8} 4×10^{-8}
$_{48}\mathrm{Cd}^{115}$ β^-, γ, e^-	(sol.)	GI (LLI)	8×10^{-8}
	(insol.)	GI (LLI)	6×10^{-8}
$_{49}\mathrm{In}^{113m}$ γ, e^-	(sol.)	GI (ULI)	3×10^{-6}
	(insol.)	GI (ULI)	2×10^{-6}
$_{49}\mathrm{In}^{114m}$ $\beta^-, \varepsilon, \gamma, e^-$	(sol.)	GI (LLI) Kidney Spleen	4×10^{-8} 4×10^{-8} 4×10^{-8}
	(insol.)	Lung GI (LLI)	7×10^{-9} 3×10^{-8}
$_{49}\mathrm{In}^{115m}$ β^-, γ, e^-	(sol.)	GI (ULI)	8×10^{-7}
	(insol.)	GI (ULI)	6×10^{-7}
$_{49}\mathrm{In}^{115}$ β^-	(sol.)	GI (LLI) Kidney	2×10^{-7} 9×10^{-8}
	(insol.)	Lung GI (LLI)	10^{-8} 2×10^{-7}
$_{50}\mathrm{Sn}^{113}$ ε, γ, e^-	(sol.)	GI (LLI) Bone	2×10^{-7} 10^{-7}
	(insol.)	Lung GI (LLI)	2×10^{-8} 10^{-7}
$_{50}\mathrm{Sn}^{125}$ β^-, γ, e^-	(sol.)	GI (LLI)	4×10^{-8}
	(insol.)	Lung GI (LLI)	3×10^{-8} 3×10^{-8}
$_{51}\mathrm{Sb}^{122}$ β^-, γ	(sol.)	GI (LLI)	6×10^{-8}
	(insol.)	GI (LLI)	5×10^{-8}
$_{51}\mathrm{Sb}^{124}$ β^-, γ	(sol.)	GI (LLI) Total body	5×10^{-8} 7×10^{-8}
	(insol.)	Lung GI (LLI)	7×10^{-9} 4×10^{-8}
$_{51}\mathrm{Sb}^{125}$ β^-, γ, e^-	(sol.)	GI (LLI) Lung Total body Bone	2×10^{-7} 2×10^{-7} 2×10^{-7} 2×10^{-7}
	(insol.)	Lung GI (LLI)	9×10^{-9} 2×10^{-7}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{52}Te^{125m}$ γ, e^-	(sol.)	Kidney GI (LLI) Testis	10^{-7} 4×10^{-7} 2×10^{-7}
	(insol.)	Lung GI (LLI)	4×10^{-8} 2×10^{-7}
$_{52}Te^{127m}$ β^-, γ, e^-	(sol.)	Kidney Testis GI (LLI)	5×10^{-8} 5×10^{-8} 2×10^{-7}
	(insol.)	Lung GI (LLI)	10^{-8} 9×10^{-8}
$_{52}Te^{127}$ β^-	(sol.)	GI (LLI)	6×10^{-7}
	(insol.)	GI (LLI)	3×10^{-7}
$_{52}Te^{129m}$ β^-, γ, e^-	(sol.)	GI (LLI) Kidney Testis	7×10^{-8} 3×10^{-8} 3×10^{-8}
	(insol.)	Lung GI (LLI)	10^{-8} 4×10^{-8}
$_{52}Te^{129}$ β^-, γ, e^-	(sol.)	GI (S)	2×10^{-6}
	(insol.)	GI (LLI)	10^{-6}
$_{52}Te^{131m}$ β^-, γ, e^-	(sol.)	GI (LLI)	10^{-7}
	(insol.)	GI (ULI)	6×10^{-8}
$_{52}Te^{132}$ β^-, γ, e^-	(sol.)	GI (LLI)	7×10^{-8}
	(insol.)	GI (LLI)	4×10^{-8}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{53}I^{126}$ β^-, ε, γ	(sol.)	Thyroid	3×10^{-9}
	(insol.)	Lung GI (LLI)	10^{-7} 2×10^{-7}
$_{53}I^{129}$ β^-, γ, e^-	(sol.)	Thyroid	6×10^{-10}
	(insol.)	Lung GI (LLI)	2×10^{-8} 4×10^{-7}
$_{53}I^{131}$ β^-, γ, e^-	(sol.)	Thyroid	3×10^{-9}
	(insol.)	GI (LLI) Lung	10^{-7} 10^{-7}
$_{53}I^{132}$ β^-, γ, e^-	(sol.)	Thyroid	8×10^{-8}
	(insol.)	GI (ULI)	3×10^{-7}
$_{53}I^{133}$ β^-, γ, e^-	(sol.)	Thyroid	10^{-8}
	(insol.)	GI (LLI)	7×10^{-8}
$_{53}I^{134}$ β^-, γ	(sol.)	Thyroid	2×10^{-7}
	(insol.)	GI (S)	10^{-6}
$_{53}I^{135}$ β^-, γ, e^-	(sol.)	Thyroid	4×10^{-8}
	(insol.)	GI (LLI)	10^{-7}
$_{54}Xe^{131m}$ γ, e^-	(submersion)	Total body	4×10^{-6}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{54}\text{Xe}^{133}$ γ, e^-	(submersion)	Total body	3×10^{-6}
$_{54}\text{Xe}^{135}$ β^-, γ	(submersion)	Total body	10^{-6}
$_{55}\text{Cs}^{131}$ ε	(sol.)	Total body Liver	4×10^{-6} 4×10^{-6}
	(insol.)	Lung GI (LLI)	10^{-6} 2×10^{-6}
$_{55}\text{Cs}^{134m}$ β^-, γ, e^-	(sol.)	GI (S)	10^{-5}
	(insol.)	GI (ULI)	2×10^{-6}
$_{55}\text{Cs}^{134}$ β^-, γ	(sol.)	Total body	10^{-8}
	(insol.)	Lung GI (LLI)	4×10^{-9} 7×10^{-8}
$_{55}\text{Cs}^{135}$ β^-	(sol.)	Liver Spleen Total body	2×10^{-7} 2×10^{-7} 2×10^{-7}
	(insol.)	GI (LLI) Lung	4×10^{-7} 3×10^{-8}
$_{55}\text{Cs}^{136}$ β^-, γ	(sol.)	Total body	10^{-7}
	(insol.)	Lung GI (LLI)	6×10^{-8} 10^{-7}
$_{55}\text{Cs}^{137}$ β^-, γ, e^-	(sol.)	Total body Liver Spleen Muscle	2×10^{-8} 3×10^{-8} 3×10^{-8} 4×10^{-8}
	(insol.)	Lung GI (LLI)	5×10^{-9} 8×10^{-8}
$_{56}\text{Ba}^{131}$ ε, γ	(sol.)	GI (LLI)	4×10^{-7}
	(insol.)	Lung GI (LLI)	10^{-7} 3×10^{-7}
$_{56}\text{Ba}^{140}$ β^-, γ	(sol.)	GI (LLI) Bone	6×10^{-8} 4×10^{-8}
	(insol.)	Lung GI (LLI)	10^{-8} 4×10^{-8}
$_{57}\text{La}^{140}$ β^-, γ	(sol.)	GI (LLI)	5×10^{-8}
	(insol.)	GI (LLI)	4×10^{-8}
$_{58}\text{Ce}^{141}$ β^-, γ	(sol.)	GI (LLI) Liver Bone	2×10^{-7} 2×10^{-7} 2×10^{-7}
	(insol.)	Lung GI (LLI)	5×10^{-8} 2×10^{-7}
$_{58}\text{Ce}^{143}$ β^-, γ	(sol.)	GI (LLI)	9×10^{-8}
	(insol.)	GI (LLI)	7×10^{-8}
$_{58}\text{Ce}^{144}$ α, β^-, γ	(sol.)	GI (LLI) Bone Liver	3×10^{-8} 3×10^{-9} 4×10^{-9}
	(insol.)	Lung GI (LLI)	2×10^{-9} 2×10^{-8}
$_{59}\text{Pr}^{142}$ β^-, γ	(sol.)	GI (LLI)	7×10^{-8}
	(insol.)	GI (LLI)	5×10^{-8}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{59}Pr^{143}$ β^-	(sol.)	GI (LLI)	10^{-7}
	(insol.)	Lung GI (LLI)	6×10^{-8} 9×10^{-8}
$_{60}Nd^{144}$ α	(sol.)	Bone GI (LLI)	3×10^{-11} 2×10^{-7}
	(insol.)	Lung GI (LLI)	10^{-10} 10^{-7}
$_{60}Nd^{147}$ α, β^-, γ	(sol.)	GI (LLI) Liver	10^{-7} 10^{-7}
	(insol.)	Lung GI (LLI)	8×10^{-8} 10^{-7}
$_{60}Nd^{149}$ β^-, γ	(sol.)	GI (LLI)	6×10^{-7}
	(insol.)	GI (ULI)	5×10^{-7}
$_{61}Pm^{147}$ α, β^-	(sol.)	GI (LLI) Bone	5×10^{-7} 2×10^{-8}
	(insol.)	Lung GI (LLI)	3×10^{-8} 4×10^{-7}
$_{61}Pm^{149}$ β^-, γ	(sol.)	GI (LLI)	10^{-7}
	(insol.)	GI (LLI)	8×10^{-8}
$_{62}Sm^{147}$ α	(sol.)	Bone GI (LLI)	2×10^{-11} 2×10^{-7}
	(insol.)	Lung GI (LLI)	9×10^{-11} 10^{-7}
$_{62}Sm^{151}$ β^-, γ	(sol.)	GI (LLI) Bone	8×10^{-7} 2×10^{-8}
	(insol.)	Lung GI (LLI)	5×10^{-8} 7×10^{-7}
$_{62}Sm^{153}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	GI (LLI)	10^{-7}
$_{63}Eu^{152}$(9.2 hr) $\beta^-, \varepsilon, \gamma$	(sol.)	GI (LLI)	10^{-7}
	(insol.)	GI (LLI)	10^{-7}
$_{63}Eu^{152}$(13 years) $\beta^-, \varepsilon, \gamma$	(sol.)	GI (LLI) Kidney	2×10^{-7} 4×10^{-9}
	(insol.)	Lung GI (LLI)	6×10^{-9} 10^{-7}
$_{63}Eu^{154}$ $\beta^-, \varepsilon, \gamma$	(sol.)	GI (LLI) Kidney Bone	5×10^{-8} 10^{-9} 10^{-9}
	(insol.)	Lung GI (LLI)	2×10^{-9} 4×10^{-8}
$_{63}Eu^{155}$ β^-, γ	(sol.)	GI (LLI) Kidney Bone	4×10^{-7} 3×10^{-8} 3×10^{-8}
	(insol.)	Lung GI (LLI)	3×10^{-8} 4×10^{-7}
$_{64}Gd^{153}$ ε, γ, e^-	(sol.)	GI (LLI) Bone	5×10^{-7} 8×10^{-8}
	(insol.)	Lung GI (LLI)	3×10^{-8} 4×10^{-7}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)	Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{64}\mathrm{Gd}^{159}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}	$_{69}\mathrm{Tm}^{170}$ β^-, ε, γ, e^-	(sol.)	GI (LLI) Bone	10^{-7} 10^{-8}
	(insol.)	GI (LLI)	10^{-7}		(insol.)	Lung GI (LLI)	10^{-8} 8×10^{-8}
$_{65}\mathrm{Tb}^{160}$ β^-, γ	(sol.)	GI (LLI) Bone Kidney Total body	10^{-7} 3×10^{-8} 4×10^{-8} 5×10^{-8}	$_{69}\mathrm{Tm}^{171}$ β^-	(sol.)	GI (LLI) Bone	10^{-6} 4×10^{-8}
	(insol.)	Lung GI (LLI)	10^{-8} 8×10^{-8}		(insol.)	Lung GI (LLI)	8×10^{-8} 9×10^{-7}
$_{66}\mathrm{Dy}^{165}$ β^-, γ	(sol.)	GI (ULI)	9×10^{-7}	$_{70}\mathrm{Yb}^{175}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	GI (ULI)	7×10^{-7}		(insol.)	GI (LLI)	2×10^{-7}
$_{66}\mathrm{Dy}^{166}$ β^-, γ, e^-	(sol.)	GI (LLI)	8×10^{-8}	$_{71}\mathrm{Lu}^{177}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	GI (LLI)	7×10^{-8}		(insol.)	GI (LLI) Lung	2×10^{-7} 2×10^{-7}
$_{67}\mathrm{Ho}^{166}$ β^-, γ, e^-	(sol.)	GI (LLI)	7×10^{-8}	$_{72}\mathrm{Hf}^{181}$ β^-, γ	(sol.)	GI (LLI) Spleen	2×10^{-7} 10^{-8}
	(insol.)	GI (LLI)	6×10^{-8}		(insol.)	Lung GI (LLI)	3×10^{8} 10^{-7}
$_{68}\mathrm{Er}^{169}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}	$_{73}\mathrm{Ta}^{182}$ β^-, γ	(sol.)	GI (LLI) Liver	9×10^{-8} 10^{-8}
	(insol.)	Lung GI (LLI)	10^{-7} 2×10^{-7}		(insol.)	Lung GI (LLI)	7×10^{-9} 7×10^{-8}
$_{68}\mathrm{Er}^{171}$ β^-, γ, e^-	(sol.)	GI (ULI)	2×10^{-7}	$_{74}\mathrm{W}^{181}$ ε, γ	(sol.)	GI (LLI)	8×10^{-7}
	(insol.)	GI (ULI)	2×10^{-7}		(insol.)	Lung GI (LLI)	4×10^{-8} 6×10^{-7}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{74}W^{185}$ β^-	(sol.)	GI (LLI)	3×10^{-7}
	(insol.)	Lung GI (LLI)	4×10^{-8} 2×10^{-7}
$_{74}W^{187}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	GI (LLI)	10^{-7}
$_{75}Re^{183}$ ε, γ	(sol.)	GI (LLI) Total body	10^{-6} 9×10^{-7}
	(insol.)	Lung GI (LLI)	5×10^{-8} 5×10^{-7}
$_{75}Re^{186}$ β^-, γ	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	GI (LLI)	8×10^{-8}
$_{75}Re^{187}$ β^-	(sol.)	GI (LLI) Skin	6×10^{-6} 3×10^{-6}
	(insol.)	Lung GI (LLI)	2×10^{-7} 2×10^{-6}
$_{75}Re^{188}$ β^-, γ	(sol.)	GI (LLI)	10^{-7}
	(insol.)	GI (LLI)	6×10^{-8}
$_{76}Os^{185}$ ε, γ, e^-	(sol.)	GI (LLI)	2×10^{-7}
	(insol.)	Lung GI (LLI)	2×10^{-8} 10^{-7}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{76}Os^{191m}$ β^-, γ, e^-	(sol.)	GI (LLI)	6×10^{-6}
	(insol.)	Lung GI (LLI)	3×10^{-6} 4×10^{-6}
$_{76}Os^{191}$ β^-, γ, e^-	(sol.)	GI (LLI)	4×10^{-7}
	(insol.)	Lung GI (LLI)	10^{-7} 3×10^{-7}
$_{76}Os^{193}$ β^-	(sol.)	GI (LLI)	10^{-7}
	(insol.)	GI (LLI)	9×10^{-8}
$_{77}Ir^{190}$ ε, γ	(sol.)	GI (LLI)	4×10^{-7}
	(insol.)	Lung GI (LLI)	10^{-7} 3×10^{-7}
$_{77}Ir^{192}$ β^-, γ	(sol.)	GI (LLI) Kidney Spleen	9×10^{-8} 4×10^{-8} 5×10^{-8}
	(insol.)	Lung GI (LLI)	9×10^{-9} 6×10^{-8}
$_{77}Ir^{194}$ β^-	(sol.)	GI (LLI)	8×10^{-8}
	(insol.)	GI (LLI)	5×10^{-8}
$_{78}Pt^{191}$ ε, γ	(sol.)	GI (LLI)	3×10^{-7}
	(insol.)	GI (LLI)	2×10^{-7}

Radionuclide and type of decay	Critical organ	Maximum permissible concentration in air (μc/ml)
$_{78}Pt^{193m}$ (sol.) ε, γ	GI (LLI)	2×10^{-6}
(insol.)	GI (LLI) Lung	2×10^{-6} 2×10^{-6}
$_{78}Pt^{193}$ (sol.) ε	Kidney	4×10^{-7}
(insol.)	Lung GI (LLI)	10^{-7} 3×10^{-6}
$_{78}Pt^{197m}$ (sol.) β⁻, γ, e⁻	GI (ULI)	2×10^{-6}
(insol.)	GI (ULI)	2×10^{-6}
$_{78}Pt^{197}$ (sol.) β⁻, γ	GI (LLI)	3×10^{-7}
(insol.)	GI (LLI)	2×10^{-7}
$_{79}Au^{196}$ (sol.) β⁻, γ, e⁻	GI (LLI)	4×10^{-7}
(insol.)	Lung GI (LLI)	2×10^{-7} 3×10^{-7}
$_{79}Au^{198}$ (sol.) β⁻, γ	GI (LLI)	10^{-7}
(insol.)	GI (LLI)	8×10^{-8}
$_{79}Au^{199}$ (sol.) β⁻, γ	GI (LLI)	4×10^{-7}
(insol.)	GI (LLI)	3×10^{-7}
$_{80}Hg^{197m}$ (sol.) ε, γ, e⁻	Kidney	3×10^{-7}
(insol.)	GI (LLI)	3×10^{-7}
$_{80}Hg^{197}$ (sol.) ε, γ, e⁻	Kidney	4×10^{-7}
(insol.)	GI (LLI)	9×10^{-7}
$_{80}Hg^{203}$ (sol.) β⁻, γ, e⁻	Kidney	2×10^{-8}
(insol.)	Lung GI (LLI)	4×10^{-8} 2×10^{-7}
$_{81}Tl^{200}$ (sol.) ε, γ	GI (LLI)	9×10^{-7}
(insol.)	GI (LLI)	4×10^{-7}
$_{81}Tl^{201}$ (sol.) ε, γ, e⁻	GI (LLI)	7×10^{-7}
(insol.)	GI (LLI)	3×10^{-7}
$_{81}Tl^{202}$ (sol.) ε, γ, e⁻	GI (LLI)	3×10^{-7}
(insol.)	Lung GI (LLI)	8×10^{-9} 10^{-7}
$_{81}Tl^{204}$ (sol.) β⁻	GI (LLI) Kidney	2×10^{-7} 2×10^{-7}
(insol.)	Lung GI (LLI)	9×10^{-9} 10^{-7}

Radionuclide and type of decay	Critical organ	Maximum permissible concentration in air ($\mu c/ml$)
$_{82}Pb^{203}$ (sol.) ε, γ	GI (LLI)	9×10^{-7}
(insol.)	GI (LLI)	6×10^{-7}
$_{82}Pb^{210}$ (sol.) α, β^-, γ	Kidney Total body	4×10^{-11} 4×10^{-10}
(insol.)	Lung GI (LLI)	8×10^{-11} 3×10^{-7}
$_{82}Pb^{212}$ (sol.) $\alpha, \beta^-, \gamma, e^-$	Kidney GI (LLI)	6×10^{-9} 4×10^{-8}
(insol.)	Lung GI (LLI)	7×10^{-9} 3×10^{-8}
$_{83}Bi^{206}$ (sol.) ε, γ	GI (LLI) Kidney	8×10^{-8} 6×10^{-8}
(insol.)	Lung GI (LLI)	5×10^{-8} 7×10^{-8}
$_{83}Bi^{207}$ (sol.) ε, γ	GI (LLI) Kidney	10^{-7} 6×10^{-8}
(insol.)	Lung GI (LLI)	5×10^{-9} 10^{-7}
$_{83}Bi^{210}$ (sol.) α, β^-	GI (LLI) Kidney	9×10^{-8} 2×10^{-9}
(insol.)	Lung GI (LLI)	2×10^{-9} 7×10^{-8}
$_{83}Bi^{212}$ (sol.) α, β^-, γ	GI (S) Kidney	8×10^{-7} 3×10^{-8}
(insol.)	Lung GI (S)	7×10^{-8} 6×10^{-7}
$_{84}Po^{210}$ (sol.) α	Spleen Kidney	2×10^{-10} 2×10^{-10}
(insol.)	Lung GI (LLI)	7×10^{-11} 5×10^{-8}
$_{85}At^{211}$ (sol.) $\alpha, \varepsilon, \gamma$	Thyroid Ovary	2×10^{-9} 3×10^{-9}
(insol.)	Lung GI (ULI)	10^{-8} 10^{-7}
$_{86}Rn^{220}$ * $\alpha, \beta^-, \gamma, e^-$	Lung	10^{-7} *
$_{86}Rn^{222}$ * α, β^-, γ	Lung	10^{-8} *
$_{88}Ra^{223}$ (sol.) α, β^-, γ	Bone	6×10^{-10}
(insol.)	Lung GI (LLI)	8×10^{-11} 7×10^{-9}
$_{88}Ra^{224}$ (sol.) $\alpha, \beta^-, \gamma, e^-$	Bone	2×10^{-9}
(insol.)	Lung GI (LLI)	2×10^{-10} 9×10^{-9}

* The daughter elements of Rn^{220} and Rn^{222} are assumed present to the extent they occur in unfiltered air. For all other isotopes the daughter elements are not considered as part of the intake and if present they must be considered on the basis of the rules for mixtures.

Radionuclide and type of decay	Critical organ	Maximum permissible concentration in air ($\mu c/ml$)
$_{88}\mathrm{Ra}^{226}$ (sol.) α, β^-, γ	Bone	10^{-11}
(insol.)	GI (LLI)	6×10^{-8}
$_{88}\mathrm{Ra}^{228}$ (sol.) $\alpha, \beta^-, \gamma, e^-$	Bone	2×10^{-11}
(insol.)	Lung GI (LLI)	10^{-11} 4×10^{-8}
$_{89}\mathrm{Ac}^{227}$ (sol.) α, β^-, γ	Bone	8×10^{-13}
(insol.)	Lung GI (LLI)	9×10^{-12} 5×10^{-7}
$_{89}\mathrm{Ac}^{228}$ (sol.) $\alpha, \beta^-, \gamma, e^-$	GI (ULI) Bone Liver	2×10^{-7} 3×10^{-8} 3×10^{-8}
(insol.)	Lung GI (ULI)	6×10^{-9} 2×10^{-7}
$_{90}\mathrm{Th}^{227}$ (sol.) α, β^-, γ	GI (LLI) Bone	$4 \times 10^{-8'}$ 10^{-10}
(insol.)	Lung GI (LLI)	6×10^{-11} 3×10^{-8}
$_{90}\mathrm{Th}^{228}$ (sol.) $\alpha, \beta^-, \gamma, e^-$	Bone	3×10^{-12}
(insol.)	Lung GI (LLI)	2×10^{-12} 2×10^{-8}
$_{90}\mathrm{Th}^{230}$ (sol.) α, γ	Bone	8×10^{-13}
(insol.)	Lung GI (LLI)	3×10^{-12} 6×10^{-8}
$_{90}\mathrm{Th}^{231}$ (sol.) α, β^-, γ	GI (LLI)	5×10^{-7}
(insol.)	GI (LLI)	4×10^{-7}
$_{90}\mathrm{Th}^{232}$ * (sol.) $\alpha, \beta^-, \gamma, e^-$	Bone	7×10^{-13}*
(insol.)	Lung GI (LLI)	4×10^{-12} 7×10^{-8}
$_{90}\mathrm{Th}^{234}$ (sol.) β^-, γ	GI (LLI) Bone	4×10^{-8} 2×10^{-8}
(insol.)	Lung GI (LLI)	10^{-8} 3×10^{-8}
$_{90}$Th-Nat * (sol.) $\alpha, \beta^-, \gamma, e^-$	Bone	6×10^{-13}*
(insol.)	Lung GI (LLI)	10^{-12} 2×10^{-8}
$_{91}\mathrm{Pa}^{230}$ (sol.) $\alpha, \beta^-, \varepsilon, \gamma$	GI (LLI) Bone	5×10^{-7} 6×10^{-10}
(insol.)	Lung GI (LLI)	3×10^{-10} 4×10^{-7}

* Provisional values for Th^{232} and Th-nat. Although calculations and animal experiments suggest that Th-nat, if injected intravenously, is perhaps as hazardous as Pu and indicate the values listed above, experience to date has suggested that in industrial circumstances the hazard of Th-nat is not much greater than that of U-nat. Therefore, pending further investigation the values $(\mathrm{MPC})_\alpha = 3 \times 10^{-11}$ $\mu c/cm^3$ for the 40-hour week and $(\mathrm{MPC})_\alpha = 10^{-11}$ $\mu c/cm^3$ for continuous occupational exposure (168 hr/wk) are recommended as provisional levels, permissible for exposure to inhaled Th-nat or Th^{232}. However, the values given in this annex are listed to indicate the possibility that further evidence may require lower values and to urge especially that exposure levels for these radionuclides be kept as low as is operationally possible. It may be possible to show that similar considerations apply to other inhaled long-lived thorium isotopes under conditions in which the physical characteristics of the airborne particulates are much the same as in the case of Th-nat, and where there is a large amount of airborne material serving as an effective carrier for the thorium.

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)	Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air (μc/ml)
$_{91}\mathrm{Pa}^{231}$ α, β^-, γ	(sol.)	Bone	4×10^{-13}	$_{92}\mathrm{U}^{236}$ α, γ	(sol.)	GI (LLI) Bone	7×10^{-8} 2×10^{-10}
	(insol.)	Lung GI (LLI)	4×10^{-11} 5×10^{-8}		(insol.)	Lung GI (LLI)	4×10^{-11} 6×10^{-8}
$_{91}\mathrm{Pa}^{233}$ β^-, γ	(sol.)	GI (LLI) Kidney	3×10^{-7} 2×10^{-7}	$_{92}\mathrm{U}^{238}$ α, γ, e^-	(sol.)	GI (LLI) Kidney	8×10^{-8} 3×10^{-11}
	(insol.)	Lung GI (LLI)	6×10^{-8} 2×10^{-7}		(insol.)	Lung GI (LLI)	5×10^{-11} 6×10^{-8}
$_{92}\mathrm{U}^{230}$ α, β^-, γ	(sol.)	GI (LLI) Kidney	10^{-8} 10^{-10}	$_{92}$U-Natural $\alpha, \beta^-, \gamma, e^-$	(sol.)	GI (LLI) Kidney	4×10^{-8} 3×10^{-11}
	(insol.)	Lung GI (LLI)	4×10^{-11} 8×10^{-9}		(insol.)	Lung GI (LLI)	2×10^{-11} 3×10^{-8}
$_{92}\mathrm{U}^{232}$ $\alpha, \beta^-, \gamma, e^-$	(sol.)	GI (LLI) Bone	6×10^{-8} 3×10^{-11}	$_{93}\mathrm{Np}^{237}$ α, β^-, γ	(sol.)	Bone	10^{-12}
	(insol.)	Lung GI (LLI)	9×10^{-12} 5×10^{-8}		(insol.)	Lung GI (LLI)	4×10^{-11} 5×10^{-8}
$_{92}\mathrm{U}^{233}$ α, γ	(sol.)	GI (LLI) Bone	7×10^{-8} 2×10^{-10}	$_{93}\mathrm{Np}^{239}$ α, β^-, γ	(sol.)	GI (LLI)	3×10^{-7}
	(insol.)	Lung GI (LLI)	4×10^{-11} 6×10^{-8}		(insol.)	GI (LLI)	2×10^{-7}
$_{92}\mathrm{U}^{234}$ α, γ	(sol.)	GI (LLI) Bone	7×10^{-8} 2×10^{-10}	$_{94}\mathrm{Pu}^{238}$ α, γ	(sol.)	Bone	7×10^{-13}
	(insol.)	Lung GI (LLI)	4×10^{-11} 6×10^{-8}		(insol.)	Lung GI (LLI)	10^{-11} 5×10^{-8}
$_{92}\mathrm{U}^{235}$ α, β^-, γ	(sol.)	GI (LLI) Kidney Bone	6×10^{-8} 2×10^{-10} 2×10^{-10}	$_{94}\mathrm{Pu}^{239}$ α, γ	(sol.)	Bone	6×10^{-13}
	(insol.)	Lung GI (LLI)	4×10^{-11} 5×10^{-8}		(insol.)	Lung GI (LLI)	10^{-11} 5×10^{-8}

Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air ($\mu c/ml$)	Radionuclide and type of decay		Critical organ	Maximum permissible concentration in air ($\mu c/ml$)
$_{94}Pu^{240}$ α, γ	(sol.)	Bone	6×10^{-13}	$_{96}Cm^{244}$ α, γ	(sol.)	Bone	3×10^{-12}
	(insol.)	Lung GI (LLI)	10^{-11} 5×10^{-8}		(insol.)	Lung GI (LLI)	3×10^{-11} 5×10^{-8}
$_{92}Pu^{241}$ α, β⁻, γ	(sol.)	Bone	3×10^{-11}	$_{96}Cm^{245}$ α, β⁻, γ	(sol.)	Bone	2×10^{-12}
	(insol.)	Lung GI (LLI)	10^{-8} 2×10^{-6}		(insol.)	Lung GI (LLI)	4×10^{-11} 5×10^{-8}
$_{94}Pu^{242}$ α	(sol.)	Bone	6×10^{-13}	$_{96}Cm^{246}$ α	(sol.)	Bone	2×10^{-12}
	(insol.)	Lung GI (LLI)	10^{-11} 5×10^{-8}		(insol.)	Lung GI (LLI)	4×10^{-11} 5×10^{-8}
$_{95}Am^{241}$ α, γ	(sol.)	Kidney Bone	2×10^{-12} 2×10^{-12}	$_{97}Bk^{249}$ α, β⁻, γ	(sol.)	GI (LLI) Bone	10^{-6} 3×10^{-10}
	(insol.)	Lung GI (LLI)	4×10^{-11} 5×10^{-8}		(insol.)	Lung GI (LLI)	4×10^{-8} 10^{-6}
$_{95}Am^{243}$ α, β⁻, γ	(sol.)	Bone Kidney	2×10^{-12} 2×10^{-12}	$_{98}Cf^{249}$ α, γ	(sol.)	Bone	5×10^{-13}
	(insol.)	Lung GI (LLI)	4×10^{-11} 5×10^{-8}		(insol.)	Lung GI (LLI)	3×10^{-11} 4×10^{-8}
$_{96}Cm^{242}$ α, γ	(sol.)	GI (LLI) Liver	5×10^{-8} 4×10^{-11}	$_{98}C^{250}$ α	(sol.)	Bone	2×10^{-12}
	(insol.)	Lung GI (LLI)	6×10^{-11} 4×10^{-8}		(insol.)	Lung GI (LLI)	3×10^{-11} 4×10^{-8}
$_{96}Cm^{243}$ α, γ	(sol.)	Bone	2×10^{-12}	$_{98}Cf^{252}$ α, γ	(sol.)	GI (LLI) Bone	2×10^{-8} 2×10^{-12}
	(insol.)	Lung GI (LLI)	3×10^{-11} 4×10^{-8}		(insol.)	Lung GI (LLI)	10^{-11} 10^{-8}

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